Atomic Energy, Vol. 86, No. 2, 1999

KINETICS OF THE LEACHING OF ^{90}Sr FROM FUEL PARTICLES IN SOIL IN THE NEAR ZONE OF THE CHERNOBYL NUCLEAR POWER PLANT

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UDC 539.12.08

A method is presented for calculating the fraction of ^{90}Sr included in fuel particles in soil. Data concerning the change in forms of the occurrence of ^{90}Sr in different soils in the 30-km zone, at different distances from the Chernobyl nuclear power plant, were used to obtain the kinetic characteristics of its leaching: the first-order rate constant and the normalized rate of solution. Depending on the direction and distance from the nuclear power plant, the first-order leaching rate constant varies from $3 \cdot 10^{-5}$ to $2 \cdot 10^{-3}$ days^{-1} and the normalized rate of solution of the fuel matrix varies from $1 \cdot 10^{-5}$ to $6.1 \cdot 10^{-4}$. It was not found possible to clearly identify the influence of the distance from the nuclear power plant on the leaching rate in the northern and western sectors. In contrast, in the southerly and south-easterly directions a clear tendency was observed for the leaching rate to increase with increasing distance from the nuclear power plant.

The key process in transforming the forms of occurrence of radionuclides in natural media during the first years after the accident at the Chernobyl nuclear power plant was their leaching from the fuel particles [1]. The loss of radionuclides from fuel particles takes place as a result of their diffusion from the volume of the particle onto the surface with subsequent passage into solution when the fuel matrix dissolves [2]. The latter was experimentally confirmed by the presence of uranium in the aqueous extract from soil (black soil in the region of the Chernobyl nuclear power plant) which has been in contact with hot particles for 290 days [3]. The aim of the present work is to investigate the kinetics of the leaching of ^{90}Sr from fuel particles in the soil of the near zone of the Chernobyl nuclear power plant.

Calculation of the Fraction of ^{90}Sr Contained in Fuel Particles. Fuel particles do not dissolve in neutral salt solutions and consequently the fraction of mobile forms of radionuclides (exchange and water-soluble) in the soil of the near zone of the Chernobyl nuclear power plant was markedly lower during the first years following the accident than in similar soil in which the radionuclides had arrived in a dissolved state [4–6]. This difference can be utilized in order to make a qualitative estimate of the fraction of a radionuclide present in soil as fuel particles. For this purpose it is preferable to employ data on the forms of presence of ^{90}Sr. Sorption equilibrium of radiostrontium in soil is reached after a few days [7] and so the form in which it occurs outside fuel particles can be considered to be the equilibrium form. In addition, the steady-state fraction of exchange radiostrontium in the majority of soils is close to 100% and is only weakly dependent on the properties of the soil [8].

The fraction of ^{90}Sr occurring in fuel particles at different places in the contaminated zone can be calculated using the formula $F_t = 1 - \alpha_t/\alpha_\infty$, where F_t is the fraction of ^{90}Sr in fuel particles in the soil at time t; α_t and α_∞ are respectively the fractions of exchange ^{90}Sr at time t and in the qualitative state.

The fraction of the exchange form of the radionuclide at a given time (α_t) is determined using the method of extracting a 1N solution of ammonium acetate with a phase ratio of 1:8 and an extraction time of 1 day. The equilibrium fraction of the exchange form of ^{90}Sr (α_∞) can be found by three methods. The most accurate method evidently involves introducing a radionuclide tracer into the investigated soil, such as ^{85}Sr in solution form, and determining the ratio of the forms of its occurrence after equilibrium has been established. Another method for determining α_∞ consists of approximating it with the steady-state fraction of the exchange form of ^{90}Sr in the lower layers of the given soil in which there are

Taifun Scientific Production Enterprise. Translated from Atomnaya Énergiya, Vol. 86, No. 2, pp. 129–134, February, 1999. Original article submitted November 11, 1998.

1063-4258/99/8602-0136$22.00

TABLE 1. Equilibrium Fraction of Exchange Forms of ^{90}Sr in Soil of the 30-km Zone of the Nuclear Power Plant, Determined from the Ratio of the Forms in Lower Soil Layers

Location	Soil	Layer, cm	Year	α_∞, %
Northern Korogod	Sod–weak-podzolic	3–5	1988	87 ± 2
	>>	3–5	1989	88 ± 2
Northern Benevka	Alluvial sod–acidic	3–5	1988	80 ± 6
	>>	4–10	1989	90 ± 4
	Alluvial meadow–acidic	4–10	1989	87 ± 7
	Sod–podzolic	4–10	1991	79 ± 10
Chernobyl town	>>	3–5	1988	85 ± 5
Flood plain of the River Pripyat'	Alluvial sod–acidic	5–10	1991	74 ± 10
Kopachi town	Marsh peat gley	8–12	1989	55 ± 10
	>>	8–10	1989	78 ± 5
	Alluvial meadow–acidic	8–12	1989	89 ± 8
	Alluvial sod–acidic	8–12	1989	88 ± 11

no fuel particles possessing migration capability [1, 9]. The values of α_∞ given in Table 1 were used to calculate F_t at the places indicated.

For the first few years following the accident, when the value of α_t was a long way from the equilibrium value, a satisfactory accuracy could be achieved by taking account of an expert estimate of α_∞. For example, in the case of the sod–podzolic soil most widely present in the contaminated region, as for other acid soil having a low content of organic matter, α_∞ lies in the range from 70 to 90% [8]. Thus, the use of these types of soil as an expert estimate of $\alpha_\infty = 0.8$ does not introduce errors of more than 10% when calculating F_t during the initial stage following the accident in the 30-km zone, where $F_t > 0.5$.

Kinetics of Leaching Radionuclides from Fuel Particles. It is difficult to model the processes resulting in radionuclides leaving hot particles because of the nonuniformity of the dimensions, forms, and chemical nature of these particles. In view of this, it is advisable to make forecasts by using integrated parameters which characterize the rate of leaching of radionuclides from fuel particles in different regions of the contaminated zone. Possible parameters of this kind are the first-order rate constant k_1 (day^{-1}) or the rate of solution of the fuel matrix v (g·day^{-1}·cm^{-2}). During the destruction and solution of fuel particles, their dimensions are reduced and the relative content of radionuclides increases in the most stable particles. The former process leads to an increase in the first-order rate constant and the latter process leads to a reduction. A description of the leaching rate of radionuclides from fuel particles is also based on the assumption that these process mutually compensate each other. In this case, the reduction with time of the fraction of radionuclides occurring in the particles is described by the following equations:

$$\frac{dF_t}{dt} = -k_1 F_t$$

and consequently

$$F_t = F_0 \exp(-k_1 t),$$

where F_t and F_0 are the fractions of radionuclides in the fuel particles respectively at the time the t and in the original fallout, and t is the time elapsed since the accident.

If the particles are of roughly the same size and have the same stability against being dissolved, the leaching rate will increase with time on account of the increase in the ratio of the surface area of the particles to their mass. In this case it is preferable to employ the rate of solution v normalized to the particle surface area as the parameter.

TABLE 2. Leaching Rate Constants of ^{90}Sr and Rates of Solution of Fuel Particles in Soil in the Zone Contaminated After the Accident at the Chernobyl Nuclear Power Plant

Populated location	Soil	Distance from nuclear power plant, km	Direction	k_1 10^4, day^{-1}	$v \cdot 10^4$, day^{-1}
North-westerly direction (northern sector)					
Novoshepelichi	Sod–weak-podzolic	8	North-westerly	1.4	0.5
Benevka	Alluvial podzolic	10	North-westerly	4.0	1.2
				3.4	1.0
	Acidic			5.8	1.6
	Alluvial			4.3	1.2
	Meadow–acidic			2.8	0.9
				7.9	2.0
	Sod-podzolic			1.5	0.5
Dovlyady	Alluvial	23	North-westerly	1.1	0.4
	Sod			1.3	0.4
	Meadow			1.4	0.5
				2.8	0.9
Westerly and south-westerly direction (western sector)					
Yanov	Sod–weak-podzolic	3	Westerly	1.1	0.4
Pripyat'	Sod-podzolic	4	>>	0.3	0.1
Rechitsa	Sod–gley	14	>>	1.1	0.4
Tolstyi Les	Alluvial acidic	20	>>	0.4	0.1
Rudnya	Alluvial	23	South-westerly	0.4	0.1

By definition

$$v = -\frac{1}{S}\frac{dP_t}{dt},$$

where P_t is the mass of the particle at time t and S is its surface area. Assuming the radionuclide to be uniformly distributed in the particle volume and taking into account that

$$\frac{dP_t}{dt} = \rho S \frac{dR_t}{dt}$$

and

$$\left(\frac{F_t}{F_0}\right)^{1/3} = \frac{R_t}{R_0},$$

where R_t and R_0 are respectively the current and initial radius of the particle, ρ is the density of the fuel matrix, and F_0 is the initial fraction of the radionuclide in the fuel particles, we obtain

$$\left(\frac{F_t}{F_0}\right)^{1/3} = 1 - \left(\frac{v}{\rho R_0}\right)t. \qquad (1)$$

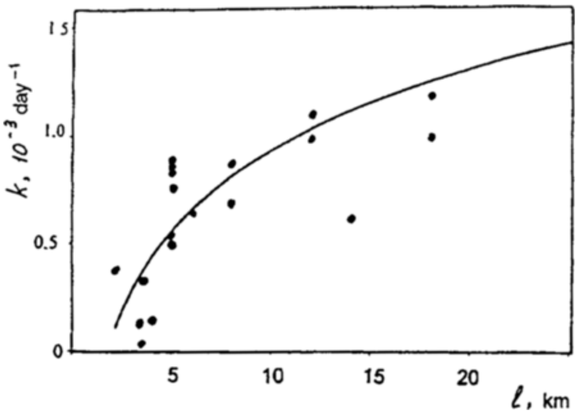

Fig. 1. Dependence of the rate constant for the leaching of ^{90}Sr from fuel particles on the distance from the Chernobyl nuclear power plant, for the southern sector.

Thus, if the particles can be characterized by one effective size and the rate of solution of the uranium matrix per unit surface area can be considered to be constant under the given conditions, then the time dependence of $(F_t/F_0)^{1/3}$ should be linear. An analog of the rate constant in this case is the normalized rate of solution $w = v/\rho R_0$.

Moreover, in the case of a description of the leaching of radionuclides in terms of the first-order kinetics, the time dependence of $\ln(F_t)$ must be linear, in accordance with the equation

$$\ln(F_t) = \ln(F_0) - kt. \quad (2)$$

Results and Discussion. The possibility of utilizing these models to predict the leaching of radionuclides was verified by comparing experimental and theoretical curves for the change with time of the ^{90}Sr content in fuel particles. This was done by utilizing the data on the content of exchange ^{90}Sr in the period 1987–1991 in the soil in the region of northern Benevka and in the experimental zone alongside northern Korogoda. The content of exchange ^{90}Sr in the initial fallout in the region of northern Korogoda is assumed to be equal to its content in the fallout onto the sample boxes of the Chernobyl weather station, namely 10% [10], and in the region of northern Benevka it was taken as 5% [11].

On a semilogarithmic scale, the time dependence of the fraction of ^{90}Sr in fuel particles should be a straight line if the leaching takes place at the rate described by Eq. (2). Moreover, if Eq. (1) is correct, then the time dependence of the ratio $(F_t/F_0)^{1/3}$ should be a straight line. The results of a regression analysis of the change of F_t/F_0 with time in accordance with Eqs. (1) and (2) showed satisfactory agreement between the experimental and theoretical dependences [12]. Here Eq. (1) gives slightly better agreement between the calculated results and the experimental data. As a rule, the correlation coefficients for Eq. (1) are higher. Evidence in favor of this equation is also given by the agreement between the rate of solution constants calculated from the change in form of the occurrence of ^{90}Sr in soil and determined in a laboratory experiment studying the action of solutions simulating natural aqueous media on fuel particles extracted from the soil in the near zone of the Chernobyl nuclear power plant. Under laboratory conditions, a rate of between $5.7 \cdot 10^{-6}$ and $1.6 \cdot 10^{-4}$ g·day^{-1}·cm^{-2} was obtained [13] from the change in form of the occurrence of ^{90}Sr for 10-μm diameter particles, $(0.1–1) \cdot 10^{-5}$ g·day^{-1}·cm^{-2}, and 100-μm diameter particles, $(0.1–1) \cdot 10^{-4}$ g·day^{-1}·cm^{-2}.

The leaching rate of radionuclides from the particles depends both on the properties of the medium in which they are present and on the characteristics of the particles themselves. It is well known that in the initial period following the accident the dispersed radioactive materials ejected from the reactor drifted in the northerly and north-westerly directions, and then to the west, south, east and north-east [12]. Since the state of the reactor changed at that time, it might have been expected that the characteristics of the fallout particles would be different in different directions from the plant. In addition, the average size and density of the particles obviously decrease with increasing distances from the source. According to the classification proposed in [14], the region contaminated after the accident can be subdivided into three sectors, namely, a western sector (north-western, western, and south-western), a southern sector (southern, south-eastern, and eastern), and a northern sector (northern and north-eastern). Table 2 gives the rate constants calculated from the data concerning the forms of occurrence of radionuclides in soils in the northern and western sectors of the trail. In these directions the influence of the distance from the station on the leaching rate constant is not tracked, in contrast with the case of the southerly and south-easterly directions where a clear tendency was observed for an increase in the leaching rate constant with increasing distance from the Chernobyl nuclear power plant (see Fig. 1). The solution rate constants behave in a similar fashion.

Since the soil covering within the 30-km zone is quite homogeneous, one can conclude that the most stable particles, as regards leaching of radionuclides, fell out in the northerly, north-westerly, westerly, and south-westerly directions from the nuclear power plant, where the leaching rate constants varied in the range from 10^{-5} to 10^{-4} day^{-1}. The rate constants increase with increasing distance from the plant in a southerly direction, and at a distance of about 20 km they reach 10^{-3} day^{-1}. It follows from this that the characteristics of the fuel particles changed significantly more abruptly in the southerly direction than in the westerly direction.

Beyond the limits of the 30-km zone, the leaching rate constants should be still greater, on account of the smaller size of the particles. For example, in the soil of the Bryansk region the first determination of the forms of occurrence was made by us in 1987. The forms of occurrence of ^{90}Sr were found to be the equilibrium forms, and this confirmed that in this region the leaching rate constant was at least $5 \cdot 10^{-3}$ day^{-1}. A similar conclusion can be drawn for the Belarus region, which is 200–250 km from the nuclear power plant, and where the forms of occurrence of ^{90}Sr in soils in 1987 were also close to the equilibrium values [6].

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