CZ9727100

DIFFUSION OF TRACE ELEMENTS AND RADIONUCLIDES IN SOILS AND CLAYS

A. Gosman, J. L. Martínez Nieto

CTU, Fac. of Nucl. Sci. & Phys. Eng., Dept. of Nuclear Chemistry
Břehová 7, 115 19 Praha 1

Key words: trace elements, radionuclides, diffusion, distribution coefficient, moisture, soils, clays, prediffusion time

The previous communications [1–2] provided some results radionuclide diffusion in soils at different moistures, and the influence of pH of soils on diffusion. Capillary modification of the thin layer method was verified. The prepared soil of a given moisture is inserted into a glass capillary, which is hermetically closed. The 6–7 days represent the standard prediffusion rest time t_k used for establishing the physico-chemical equilibrium in the system. After t_k the capillary is opened at one end, paper disc with spread radionuclide is inserted and contacted with the soil, and the capillary is immediately closed: diffusion can begin. After the chosen time t the capillary is opened, very thin layers of a thickness of approx. 0.05 cm are pressed out, and their specific activities are measured and used for calculating D.

This communication discusses the next data on ^{65}Zn,^{85}Sr, ^{137}Cs diffusion in soil (LP 26, at Lesní potok, Central Bohemia, locality of Jevany). Further experiments were extended to clays in Northern Bohemia 99 (overbed, Sokolov). It was important for the data and the methods analysis used to find out if and how the diffusion parameters D_{ef} depend on the "prediffusion rest time" – i.e. on establishing the physico-chemical equilibrium, distribution of the liquid phase, etc. Besides t_k, shaking was also used in the closed cell, and after the chosen time t_p the material was placed into the capillary (start of the diffusion time). Cheung [3] also supposed certain dependence of diffusivity on time – in this case on the diffusion time proper. However, Cho, Oscarson, Hahn [4] have proved that even tens of days (e.g. 24 to 188 days) of diffusion excerted no influence on the diffusivity.

From the results obtained it follows that for soil LP 26 with particle diameter < 0.2 cm, ^{65}Zn and near moistures (20 and 25.5 w.%) the D_{ef} values equal $2.34 \cdot 10^{-7}\,cm^2s^{-1}$ and $3.02 \cdot 10^{-7}\,cm^2s^{-1}$ corresponding to t_k = 1 week; however, for the same case but t_k = 2 weeks, the D_{ef} values are $4.17 \cdot 10^{-7}\,cm^2s^{-1}$ and $5.25 \cdot 10^{-7}\,cm^2s^{-1}$. For the same soil, particle dimensions less than 0.2 cm and greater than 0.047 cm, and near moistures for t_k = 0 day it follows that $D_{ef}(^{137}Cs) < D_{ef}(^{65}Zn) < D_{ef}(^{85}Sr)$. It corresponds to changes of distribution coefficients K_d: $K_d(^{137}Cs)$ being $\approx$ 10 times greater than $K_d(^{65}Zn)$ and $K_d(^{85}Sr)$, which are of the same order. On the other hand, for the given radionuclide and change of t_k in the same soil it was found that for ^{85}Sr and near moistures $D_{ef}(^{85}Sr)$ increases with t_k = 0, t_k = 1, t_k = 2 days (there is the maximum of D_{ef}), and with the next change of t_k = 6, t_k = 29 days the $D_{ef}(^{85}Sr)$ decreases. Similarly, the maximum was found for $D_{ef}(^{65}Zn)$ if t_k changed (0–1–2–6 days).

Of importance may be the comparison of diffusivities at near moistures and t_k = const in soil (LP 26) and in clay, where D_{ef} values are by one order of magnitude lower. In

case of clay (at near moistures and t_k = const) the following has been found: at $t_k = 0$ day is $D_{ef}(^{85}\mathrm{Sr}) > D_{ef}(^{137}\mathrm{Cs}) > D_{ef}(^{65}\mathrm{Zn})$. This corresponds with the K_d change in clay: $K_d(^{65}\mathrm{Zn}) > K_d(^{137}\mathrm{Cs}) > K_d(^{85}\mathrm{Sr})$. The result is the same for $t_k = 1$ day. The explanation consists in that the contribution of the diffusion flow in the liquid phase is greatest for minimum $K_d(^{85}\mathrm{Sr})$. As regards t_p and t_k values, it has been found that in case of ^{85}Sr the influence of t_p is greater than the influence of t_k in clay compared with soil. Further, for all experiments with ^{65}Zn, ^{85}Sr and ^{137}Cs it follows that D_{ef} values at all moistures are less than the corresponding limiting Nernst D_0 values.

Conclusions and results:

a) in soil medium diffusivity data have been completed
b) diffusivity of ^{65}Zn, ^{85}Sr, and ^{137}Cs in clay has been determined by using the checked capillary modification of the thin layer method
c) it has been found that D_{ef} values in the clay medium are by one order of magnitude lower compared with the diffusion data for soil medium
d) the influence of prediffusion time of establishing physico chemical equilibrium in moist-adsorbing medium on diffusion of trace elements and radionuclides has been verified
e) the data analysis shows the possible use of the checked and carried out experimental methods for determining the diffusivity under the given and changing conditions of diffusion transport in soils, clays and other materials.

References:

[1] GOSMAN, A. - BLAŽÍČEK, J.: *Study of Diffusion of Trace Elements and Radionuclides in Soils. Capillary Modification of the Thin Layer Method. Diffusion of ^{137}Cs.* J. Radioanl. Nucl. Chem., Articles, Vol. 182, No. 2 (1994), pp. 179–191.

[2] GOSMAN, A. - BÍLÁ, N. - LEBEDA, O. - ŠUSTROVÁ, L.: *Diffusion of Trace Elements and Radionuclides in Soils.* Workshop 96 - Part III, Czech Technical University, Prague 1996, pp. 949–950.

[3] CHEUNG, S. C.: *Methods to Measure Apparent Diffusion Coefficient in Compacted Bentonite Clays and Data Interpretation.* Can.J.Civ.Eng., Vol. 16 (1989), pp. 434–443.

[4] CHO, W. J. - OSCARSON, D. W. - HAHN, P. S.: *The Measurement of Apparent Diffusion Coefficient in Compacted Clays: an Assessment of Methods.* Applied Clay Science, Vol. 8 (1993), pp. 283–294.

This research has been conducted at the Department of Nuclear Chemistry as part of the research project "Modelling of the Migration of Toxic and Radiotoxic Substances in Soils, Rocks and Sediments" and has been supported by GAČR grant No. 104/94/1517.