J Radioanal Nucl Chem (2011) 287:247–254
DOI 10.1007/s10967-010-0691-5

Adsorption of ^{85}Kr radioactive inert gas into hardening mixtures

D. Butkus · J. Kleiza

Received: 19 June 2010 / Published online: 14 September 2010

Abstract Changes in volumetric activity of ^{85}Kr radioactive inert gas take place in the atmosphere: it has increased by around 50% during the past 15 years. The main source of such gas is the operation of nuclear power plants and spent nuclear fuel reprocessing plants. ^{85}Kr as an inert gas spreads throughout the entire atmosphere and its ionizing radiation may result in changes of atmospheric electric phenomena. Therefore it is necessary to control ^{85}Kr emission into the atmosphere. However, there is no effective method for this as inert gases, under normal conditions, can hardly be adsorbed in different adsorbents and stored in special containers for a long period of time. This paper tries to show the possibility of keeping ^{85}Kr longer within the adsorbent by changing its aggregate state: gas is adsorbed into liquid adsorbent and desorption takes place from solid adsorbent. For this purpose, an epoxy resin is used which, after adding a special hardener at room temperature, turns into a solid material with density of around 1.2×10^3 kg m^{-3}. As a result of sample blending with substances which contribute to better solubility of ^{85}Kr, diffusion coefficient of this gas (i.e. desorption speed) changes within the adsorbent in the solid state.

Keywords Krypton-85 · Volumetric activity · Adsorption · Time of desorption

D. Butkus (✉)
Department of Environment Protection, Vilnius Gediminas Technical University, Saulėtekio, 11, 10223 Vilnius, Lithuania
e-mail: butkus@vgtu.lt

J. Kleiza
Department of Mathematical Modelling, Vilnius Gediminas Technical University, Saulėtekio, 11, 10223 Vilnius, Lithuania

Introduction

As nuclear power expands, radioactive inert gases enter the atmosphere. Long-lived radioactive inert gases take part in the atmospheric circulation system and spread throughout the entire atmosphere. One of the long-lived radioactive inert gases of high importance is Krypton-85 ($^{85}_{36}$Kr). The radioactivity decay half-life of this radionuclide is $T_{1/2} =$ 10.76 m; radioactivity decay constant is $\lambda = 2.04 \times 10^{-9}$ s^{-1}; 99.563% of radiation consists of beta particles with energy of 0.687 MeV, and the rest of 0.437% of radiation −0.173 MeV. Gamma radiation energy is equal to 0.514 MeV [1, 2].

One of the sources of ^{85}Kr in the atmosphere is nuclear explosions and not only in the atmosphere but also under the earth, or under water. A 1 Mt nuclear explosion results in the fission of around 56 kilograms of ^{235}U; this means 4.6×10^{23} of ^{85}Kr atoms released (0.32% yield) [3]. Supposedly, emissions of ^{85}Kr from this source amounted to 111–185 PBq in the atmosphere in 1973 [4]. Atmospheric activity of 5×10^6 Ci caused by ^{85}Kr during the period 1945–1962 is indicated in the article Krypton-85 [1].

The other way ^{85}Kr gets into the atmosphere is during the operation of nuclear plants; in cases where ^{235}U fuel is used, activity of this gas amounts to 6.2×10^{12} Bq (MW m)$^{-1}$, ^{233}U—12.2×10^{12} Bq (MW m)$^{-1}$, ^{239}Pu—2.7×10^{13} Bq (MW m)$^{-1}$ [3]. Large volumes of ^{85}Kr have been released into the atmosphere during accidents in nuclear power plants; e.g. Chernobyl Nuclear Plant catastrophe resulted in emissions of ^{85}Kr with activity of around 35 PBq [1, 4], and the Three Mille accident—around 5×10^4 Ci (185×10^{13} Bq) [1].

The significant source of atmospheric emissions of ^{85}Kr is spent nuclear fuel reprocessing (regeneration) plants; capacity of the activity of ^{85}Kr emitted by large plants might amount to 1.8×10^9 Bq s^{-1} [2, 3]. Winger et al. [4] provided detailed evaluation of ^{85}Kr emissions from spent nuclear fuel reprocessing plants in La Hague (France), Chelyabinsk (Russia), Tomsk (Russia), Sellafield (UK), Dounray (UK), and Hanford (USA). The above mentioned authors have estimated that activity of ^{85}Kr emitted into the atmosphere during the period 1945–2000 reaches up to 10.6×10^{18} Bq [4]. The paper of Smith et al. [5] determines significant increase of ^{85}Kr activity at the sampling location which was in downwind direction from La Hague nuclear fuel reprocessing plant. Most of the authors' papers [1, 4–7] declare continuous increase of ^{85}Kr activity concentration in the atmosphere. Dubasov and Okunev [8] had been monitoring ^{85}Kr activity concentration in Cherepovets district (Vologda region) and determined that activity concentration (volumetric activity) of this radionuclide had increased almost by 50% during 15 years and grew from 1.41 to 1.94 Bq m^{-3} during the period 2006–2008. This results correspond to the data of Japanese researchers [9]. As in the past, nowadays, scientists pay much of their attention to modelling the spread of ^{85}Kr in the atmosphere as this helps not only to research the distribution of this gas in the atmosphere of the Earth, but also provides information on air mass movement [4, 10–12].

Accumulation of ^{85}Kr in the atmosphere intensifies the process of ionization and this might result in irreversible climate changes. Increase of ionization in the atmosphere due to the impact of ionizing radiation of ^{85}Kr is shown in the paper by Loosli [13] and proven in the article by Harrison and ApSimon [14]. Possible increase of lightweight ion concentration until 2015 had been researched in the work by Styra and Butkus [3]. Modelling experiments have determined the dependence of lightweight ion concentration increase under conditions of ^{85}Kr gas being in contaminated environment [15–18]. Atmospheric ionization may influence geophysical processes most significantly in the atmosphere above the Earth's surface up to thunderclouds and inside of them. Beta radiation of average energy (0.251 MeV), along the travel distance of 0.751 m, creates 6,459 ion couples in the atmosphere. Increase of ion concentration in the atmosphere results in change of aerosol particle concentration and spectrum. The question whether such processes are capable of changing electrical resistance of the atmosphere and all the atmospheric processes related to it cannot be answered yet.

The other atmospheric phenomena resulted by ionizing radiation of ^{85}Kr is the ionization effects inside the thunderclouds. Only the ^{85}Kr, being long-lived radionuclide and inactive in reactions with environmental objects, is able to achieve the same volumetric activity inside a thundercloud as on the Earth's surface [2]. Distribution of radionuclides in the atmosphere provides with information on the flow of air masse [19].

For the purpose of finding out possible impact of ionizing radiation of ^{85}Kr to the atmospheric processes, it is necessary to develop modelling experiments in that direction and compare the results with feasible natural observations. We see the necessity of the following experiments and theoretic researches:

1. Containment of ^{85}Kr to prevent its emissions into the atmosphere from nuclear power plants and nuclear fuel reprocessing companies.
2. Development of radioactive inert gas chemistry with the purpose of finding substances in which ^{85}Kr could be adsorbed or chemically combined and then disposed in radioactive material containers for a long time.
3. Experimental research of the environmental ionization caused by ionizing radiation of ^{85}Kr.
4. Development of theoretic researches of the above mentioned processes.

Adsorption of ^{85}Kr in various substances was actively researched around the middle of the twentieth century [2]. Most of the works of this nature were done using activated charcoal as an adsorbent [20]. However, effective adsorption of inert gases and long time storage in the activated charcoal or other adsorbents is only possible at low temperatures [16].

We have to admit that search for new adsorbents for ^{85}Kr is negligible during current period; in fact, some research results have not only scientific but also practical significance [21]. Sorption capacity of radioactive materials varies with an adsorbent pH, time of material contact with an adsorbat [22, 23] and physical status of an adsorbent [24]. However, data to characterize adsorption of ^{85}Kr was not found.

The objective of this work is to increase desorption duration and extend the period of gas storage in adsorbents by means of adsorption of long-lived radioactive inert gases into hardening mixtures.

Experimental

For the purpose to achieve the objective of the work, the modelling experiment has been carried out; it started on April 23, 1999. Samples (adsorbents) of six types have been prepared:

Sample 1. E-6 epoxy resin and a hardener; ratio 1:0.1. Sample volume $V = 122 \times 10^{-6}$ m^3 (a cylinder with diameter of $d = 72 \times 10^{-3}$ m and a height of $h = 3 \times 10^{-2}$ m).

Sample 2. E-6 epoxy resin, hardener and toluene; ratio 1:0.1:0.15. Sample volume 163×10^{-6} m^3 (a cylinder with diameter of $d = 72 \times 10^{-3}$ m and a height of $h = 4 \times 10^{-2}$ m).

Sample 3. E-6 epoxy resin, hardener and type BAU activated charcoal; ratio 1:0.1:0.5. Sample volume

$V = 324 \times 10^{-6}$ m^3 (a cylinder with diameter of $d = 72 \times 10^{-3}$ m and a height of $h = 55 \times 10^{-3}$ m).

Sample 4. E-6 epoxy resin, hardener and type BAU activated charcoal; ratio 1:0.1:0.35. Sample volume $V = 10^{-4}$ m^3 (a cylinder with diameter of $d = 72 \times 10^{-3}$ m and a height of $h = 24,6 \times 10^{-3}$ m).

Sample 5. E-6 epoxy resin, hardener and xylene (grade A oil xylene); ratio 1:0.1:0.35. 0.1:0.13. Sample volume $V = 122 \times 10^{-6}$ m^3 (a cylinder with diameter of $d = 72 \times 10^{-3}$ m and a height of $h = 3 \times 10^{-2}$ m).

Sample 6. E-6 epoxy resin, hardener and butanol (C_4H_9OH); ratio 1:0.1:0.13. Sample volume $V = 122 \times 10^{-6}$ m^3 (a cylinder with diameter of $d = 72 \times 10^{-3}$ m and a height of $h = 3 \times 10^{-2}$ m).

During the experiments, temperature of samples has been increasing, especially with type BAU activated charcoal. In this case, volume and temperature of the solid has been increasing rapidly and even broke the closed glass container. There was almost no temperature increase in mixture of epoxy resin and xylene.

The experiment was engineered in the following way. In the "hot zone", a glass container with volume of 0.5×10^{-3} m^3 was filled with ^{85}Kr gas and sealed. The activity of ^{85}Kr gas contained in the vessel has been measured and amounted to 2.75×10^6 Bq. Gas with such activity was dosed into the container (1) of the experimental system shown in Fig. 1.

Using radioactive gas with known activity value, the effectiveness of the counter 4 (Fig. 1) has been determined and amounted to $K_1 = 0.79 \times 10^{-3}$ imp Bq^{-1} s^{-1} for ^{85}Kr gas. It was used for determination of volumetric activity of ^{85}Kr in the system shown in Fig. 1.

While the mixture in the container (2) (Fig. 1) is not hardened, the bubbling method is used to saturate the sample

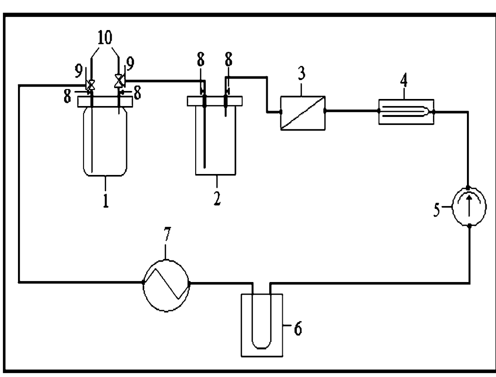

Fig. 1 Layout of experimental equipment: *1*—^{85}Kr dosing container; *2*—glass container with hardening mixture; *3*—filter; *4*—SBT-11 counter for ^{85}Kr activity measurement; *5*—reometer; *6*—column submersed into a liquid nitrogen for adsorption of ^{85}Kr contained in the system (system cleaning of ^{85}Kr); *7*—pump; *8*—valve for pressurizing containers (1) and (2); *9*—3-way valve for dosing system diversion; *10*—hoses to/from ^{85}Kr source camera; *11*—fume cupboard

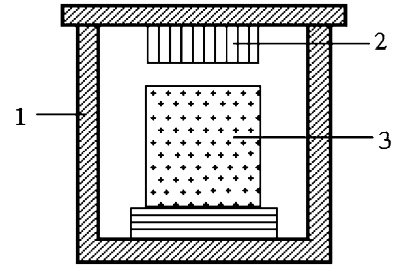

Fig. 2 Diagram of the housing for measurements of activity of ^{85}Kr radiation emitted from solid samples: *1*—lead housing; *2*—SBT-10 counter; *3*—sample

with ^{85}Kr gas. A sample is taken for determination of volumetric activity of ^{85}Kr by means of UMF-1500M low background radiation device. Effectiveness of this device, when measuring in the standard dish of the device, amounts to $K_2 = 10^{-3}$ imp Bq^{-1} s^{-1}. Volume of the mixture used to fill the dish is $V_1 = 1.96 \times 10^{-6}$ m^3. Calculation of measurement results is performed using this formula:

$$A_{V_1} = \frac{N_1}{K_2 \times V_1}, \quad (1)$$

where A_{V_1} is the volumetric activity of ^{85}Kr in a sample, Bq m^{-3}; N_1 the sample radiation intensity, imp s^{-1}; K_2 the effectiveness of the UMF-1500M device, imp Bq^{-1} s^{-1}; and V_1 is the volume of the sample measured, m^3. Activity of the background radiation equals to 0.22 imp s^{-1}.

After the mixture in container (2) (Fig. 1) hardens, circular suction in the system is stopped, containers (1) and (2) are pressurized and ^{85}Kr remaining in the system is sucked into the column (6). After this, container (2) is disconnected from the system, then broken and the activity of the cylinder-shaped sample taken out is measured using SBT-10 counter.

Activity of the radiation emitted from the surface of the solid sample taken out from the container (2) (Fig. 1) was measured by the SBT-10 counter contained within the lead housing, the diagram of which is shown in Fig. 2. At the beginning of each measurement, the background radiation was determined and fluctuated from 55 to 63 imp s^{-1}.

^{85}Kr is likely distributed evenly within the sample contained in vessel (2) and the volumetric activity of the gas is close to that of air in the system as inert gases form no compounds at normal conditions.

Results and discussion

^{85}Kr desorption from samples

Results of measured volumetric activity of ^{85}Kr in the system's (Fig. 1) air are compared with those calculated by the formula (1). Data obtained presented in Table 1.

Table 1 ^{85}Kr adsorption level in samples

Sample	Ratio of volumetric activity of ^{85}Kr in the system's (Fig. 1) air and a sample
Mixture of E-6 epoxy resin and a hardener	**1.03**
Mixture of E-6 epoxy resin, hardener, and toluene	0.30
Mixture of E-6 epoxy resin, hardener and type BAU activated charcoal (1:0.1:0.5)	0.37
Mixture of E-6 epoxy resin, hardener, and butanol	0.30
Mixture of E-6 epoxy resin, hardener, and xylene	**1.06**
Mixture of E-6 epoxy resin, hardener and type BAU activated charcoal (1:0.1:0.35)	0.33

Table 2 ^{85}Kr desorption rate

Sample No.	Mean desorption rate during 697 days, 10^{-5} m^{-2} s^{-1}	Mean desorption rate during 3,421 days, 10^{-5} m^{-2} s^{-1}	Mean desorption rate during 3,461 days, 10^{-5} m^{-2} s^{-1}
1	5.5	–	2.1
2	5.3	–	1.9
3	7.5	–	–
4	7.5	2.4	–
5	4.2	2.1	–
6	6.0	2.1	–

As we can see in the Table 1, accumulation of ^{85}Kr radioactive inert gas in respect to environmental ^{85}Kr can only be expected in pure E-6 epoxy resin and mixture with xylene.

Hardened samples were stored at room temperature and a proportion of desorpted ^{85}Kr was measured at certain intervals using equipment shown in Fig. 2. Results of the analysis presented in Table 2.

The most intense desorption takes place during the first 100 days: it amounts to 1.91×10^{-4} m^{-2} s^{-1} from mixture of E-6 epoxy resin, hardener and toluene, and mixture of E-6 epoxy resin and a hardener. Desorption significantly slows down in 200 days. ^{85}Kr desorption rate from mixture of E-6 epoxy resin, hardener and toluene, and mixture of E-6 epoxy resin and a hardener, not taking into account the decay of nuclear isotope, amounts to an average of 1.9×10^{-5} m^{-2} s^{-1} and 2.1×10^{-5} m^{-2} s^{-1} accordingly.

After all samples were stored 3,870 days at room temperature and then cut horizontally, density of the surface activity (Bq m^{-2}) of ^{85}Kr in the middle layer was significantly higher than the surface layer. This data presented in Table 3.

Table 3 Ratio of ^{85}Kr surface activity densities

Sample	Ratio of ^{85}Kr surface activity densities in the inner and outer layers
1	3.1
2	9.3
3	1.0
4	7.2
5	5.2
6	3.8

Figure 3 shows change of ^{85}Kr activity concentration (%) in Sample 6 from the centre towards the edge depending on the time t (days).

Model of Krypton concentration distribution in a sample

Decrease of concentration of any radioactive substance in a sample is influenced by two interdependent physical phenomena: spread (diffusion) of a substance through the surface of a sample according to the diffusion equation:

$$\frac{\partial C_1}{\partial t} = D\Delta C_1,$$

and radioactive decay, intensity of which is proportional to the residual volume of a substance or the activity concentration:

$$\frac{\partial C_2}{\partial t} = -\frac{\ln 2}{T_{1/2}} C_2,$$

where D is the diffusion coefficient, Δ the Laplace operator, $T_{1/2}$ the decay half-life, and C_1, C_2 is the respective concentrations.

As these phenomena take place simultaneously, total distribution of concentration in a cylinder-shaped sample (R—base radius, H—height) in the polar coordinate system (φ, r, z) might be defined by the following boundary value problem:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial z^2}\right) - \frac{\ln 2}{T_{1/2}} C \quad (2)$$

$$C|_{r=R} = C|_{z=0} = C|_{z=H} = 0. \quad (3)$$

Due to the axial symmetry of the sample shape, it is assumed that concentration is not dependent on the angle φ. Moreover, it is supposed that activity concentration, at the beginning of the experiment, is the same in each point of a sample:

$$C|_{t=0} = C_0 = \text{const}. \quad (4)$$

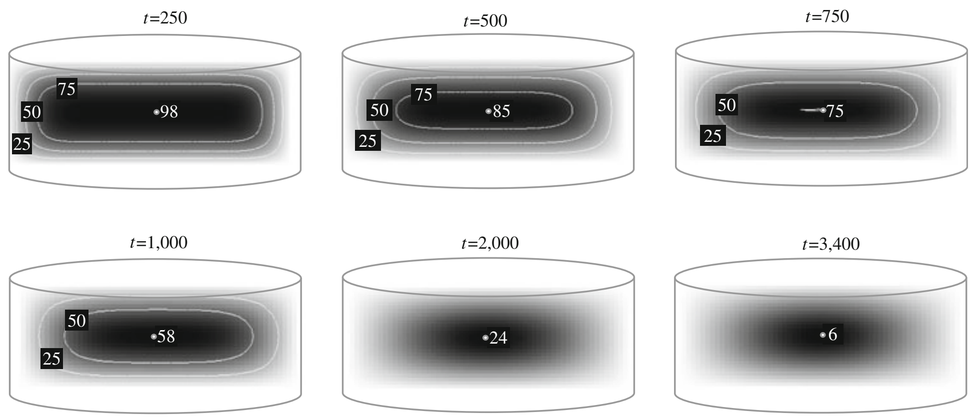

Fig. 3 Change of ^{85}Kr activity concentrations (%) in Sample 6 depending on the time t (days)

Solution of the marginal problem for distribution of activity concentration of Krypton

Traditionally, boundary value problems in mathematical physics are solved by approximate methods; however, in case of a cylinder-shaped sample, the diffusion problem (2)–(3) can be expressed in analytical form using the method of separation of variables [25]:

$$C(r,z,t) = \sum_{i=1}^{\infty}\sum_{j=1}^{\infty} a_{i,j} J_0(\mu_j r/R)\sin(i\pi z/H)\exp(-b_{i,j}t),$$

where

$$a_{i,j} = \frac{4}{R^2 H J(\mu_j)} \int_0^H \int_0^R C_0(r,z) J_0(\mu_j r/R)\sin(i\pi z/H) r\mathrm{d}r\mathrm{d}z \tag{5}$$

$$b_{ij} = D\left((\mu_j/R)^2 + (i\pi/H) + \ln 2/T_{1/2}\right),$$

where $C_0(r,z)$ is the initial distribution of concentrations; $J_k(r)$ the Bessel functions of order k; and μ_j is the roots of equation $J_0(\mu) = 0$.

On the grounds of condition (4), integral (5) could be calculated and the result is:

$$a_{2i-1,j} = 8C_0/(\pi(2i-1)\mu_j),\ a_{2i,j} = 0$$

Solution of the problem (2)–(3) might be expressed as a product of two separate series:

$$\begin{aligned} C(r,z,t) &= \frac{8C_0}{\pi}\exp(-t\ln 2/T_{1/2}) \\ &\times \sum_{i=1}^{\infty}\frac{\sin(\pi(2i-1)z/H}{2i-1}\exp\left(-Dt(\pi(2i-1)/H)^2\right) \\ &\times \sum_{j=1}^{\infty}\frac{J_0(\mu_j r/R)}{\mu_j J_1(\mu_j)}\exp\left(-Dt(\mu_j/R)^2\right), \\ &0 \le r \le R,\ 0 \le z \le H. \end{aligned} \tag{6}$$

Calculation of diffusion coefficient

Let's take $Q(t)$ as an amount of diffused ^{85}Kr during time period of $(0,t)$. Then, the result of integration of flow $-D\int_S \frac{\partial C}{\partial n}\mathrm{d}s$ according to time is as follows:

$$\begin{aligned} Q(t) &= \int_0^t -D\int_S \frac{\partial C}{\partial n}\mathrm{d}s\mathrm{d}t = -D\int_0^t\int_V \Delta C\mathrm{d}V\mathrm{d}t \\ &= -D\int_0^t\int_0^{2\pi}\mathrm{d}\varphi\int_0^H\int_0^R\left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial z^2}\right) r\mathrm{d}r\mathrm{d}z\mathrm{d}t, \end{aligned} \tag{7}$$

where S,V is the sample's surface area and volume accordingly and Δ is the Laplace operator.

The initial amount of ^{85}Kr is $Q_0 = \pi R^2 H C_0$ so calculation of integrals (7) including (6) results in the following expression of value $Q(t)/Q_0$ characterizing the desorption rate:

$$\begin{aligned} \frac{Q(t)}{Q_0} &= 1 - \frac{32}{\pi^2}\exp(-t\ln 2/T_{1/2}) \\ &\times \sum_{i=1}^{\infty}\frac{\exp\left(-Dt(\pi(2i-1)/H)^2\right)}{(2i-1)^2}\sum_{j=1}^{\infty}\frac{\exp\left(-Dt(\mu_j/R)^2\right)}{\mu_j^2}. \end{aligned} \tag{8}$$

Table 4 Mean values of diffusion coefficients of ^{85}Kr

Sample	Diffusion coefficient D, $\times 10^{-13}$ m^2s^{-1}
1	3.77
2	6.67
3	130.79
4	14.0
5	3.55
6	7.84

Series of the obtained expression converge rapidly enough (not slower than $1/i^2$ and $1/j^2$) therefore, after calculation of the sum of several tens of members, the accuracy achieved is not lower than the accuracy of desorption measurements performed. Least-squares method (i.e. comparison of calculated $Q(t)/Q_0$ values having volatile diffusion coefficient with measured $\widetilde{Q}(t_i)/Q_0$ values) is used to find approximate values of diffusion coefficients of corresponding samples. These values are presented in Table 4 and distribution of measured $\widetilde{Q}(t_i)/Q_0$ values in respect of curves (8) is shown in Fig. 4.

Mean-square error values, calculated using expression

$$d = \frac{\sqrt{\sum_{i=1}^{n}\left(Q(t_i)/Q_0 - \widetilde{Q}(t_i)/Q_0\right)}}{n}100\%,$$

stay below 2% in all samples (vary from 0.67 to 2%), and this allows us to suppose that the mathematical model used

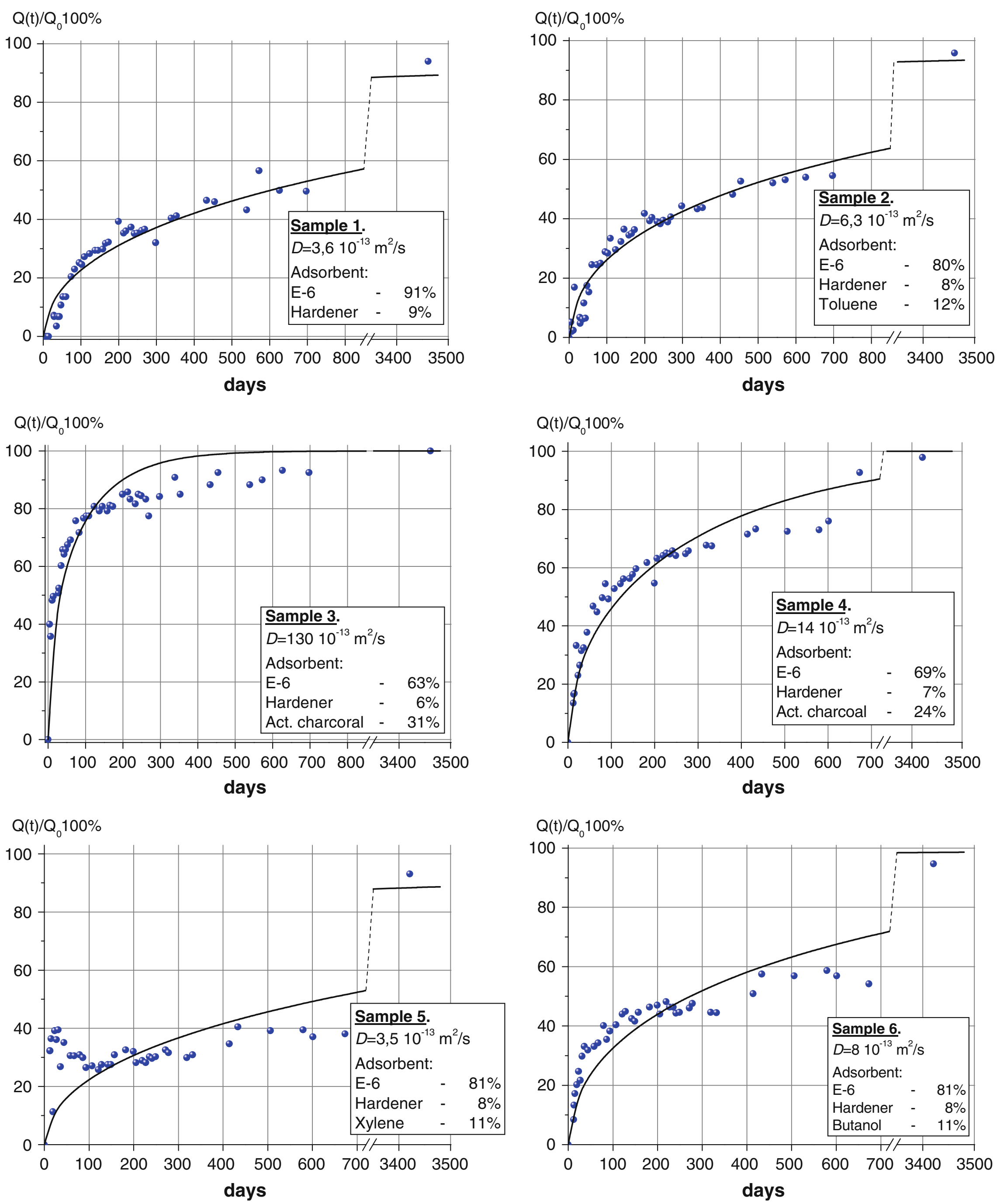

Fig. 4 Measurement results and approximation curves of the data (8)

corresponds sufficiently accurately to the process of activity concentration change, and the values of diffusion coefficients calculated might be used for evaluation of adsorption features of various aggregates.

Conclusions

Cylinder shaped, E-6 epoxy resin based adsorbents with additions of hardener, xylene, toluene, butanol, and activated carbon have been made; volume of the cylinders was 1×10^{-4} m^3 to 3.24×10^{-4} m^3 and density in solid state amounted to around 1.2×10^3 kg m^{-3}.

In different samples, the volumetric activity of ^{85}Kr adsorbed into a liquid sample varied from 0.3×10^9 Bq m^{-3} (Sample 3: mixture with charcoal) to 3.8×10^9 Bq m^{-3} (Sample 5: mixture with xylene).

Desorption of ^{85}Kr has been registered during 3,461 days, during 697 of which this was performed regularly. During the mentioned period, when desorption of ^{85}Kr has been taking place at room temperature, the mean diffusion coefficient was determined in the adsorbent.

Sample 1—3.77×10^{-13} m^{2}s^{-1}; Sample 2—6.67×10^{-13} m^{2}s^{-1}; Sample 3—130.79×10^{-13} m^{2}s^{-1}; Sample 4—14.00×10^{-13} m^{2}s^{-1}; Sample 5—3.55×10^{-13} m^{2}s^{-1}; Sample 6—7.84×10^{-13} m^{2}s^{-1}.

As diffusion of ^{85}Kr from a sample into the environment takes place, the distribution of ^{85}Kr in not continuous in a sample; after 3,870 days, the density of the surface activity of ^{85}Kr is higher in outer layers than in the inner ones: Sample 1—by 3.1 times; Sample 2—by 9.3 times; Sample 4—by 7.2 times; Sample 5—by 5.2 times; Sample 6—by 3.8 times.

Adsorption effectiveness and desorption rate of ^{85}Kr in a sample depends on sample's admixtures which contribute to rapid solubility of this gas.

Acknowledgments Experimental research has been started at the Institute of Physics. Authors express their sincere thanks to the administration of the Institute of Physics for the material facilities, and also senior engineer Gintas Kandrotas and technician Jurijus Podoroga for their contribution in preparing the samples and conducting the research.

References

1. Krypton-85. http://en.wikipedia.org/wiki/krypton-85.2009
2. Butkus D (1999) Radioactive inert gases of the artificial origin in the environment: scientific investigations and technical solutions. Summary of the research report presented for habilitation, 47 pp
3. Styra B, Butkus D (1990) Geophysical problems of Krypton-85 in the atmosphere. Hemisphere Publishing Corporation, New York, 153 pp
4. Winger K, Feichter J, Kalinowski MB, Sartorius H, Schlosser C (2005) A new compilation of the atmospheric Krypton-85 inventories from 1945 to 2000 and its evaluation in a global transport model. J Environ Radioact 80:183–215
5. Smith K, Murray M, Wong J, Long SC, Colgan PA, Rafferty B (2005) Krypton-85 and other airborne radioactivity measurements throughout Ireland. Radioprotection 40(1):5457–5463. doi: 10.1051/radiopro:2005s1-067
6. Вилгелмова Л, Томашек М, Дворжак З, Буткус Д, Земкаюс К, Стыро Б (1991) Определение атмосферных концентраций ^{85}Kr в Праге и в Вильнюсе [Vilgelmova L, Tomashek M, Dvorzhak Z, Butkus D, Zemkayus K, Styro B (1991) Determination of atmospheric concentrations of ^{85}Kr in Prague and Vilnius]. Физика атмосферы [Atmos Phys] 15:21–29
7. Achkasov SK, Gudkov AN, Zakharov OV, Krylov AYu, Nekrasov VM, Novichkov VP, Serbulov YuA, Ushakova NP, Zadorozhnyj YuA (1991) Monitoring the contamination of the atmosphere by ^{85}Kr. Atomnaya Energiya 70(4):234–239
8. Dubasov YV, Okunev NS (2010) Xenon and Krypton-85 radionuclides monitoring in the northwest region. Pure Appl Geophys 167(4–5):487–498
9. Mamoshima N, Inoue F, Sugihara S, Shimada J, Taniguchi M (2010) An improved method for ^{85}Kr analysis by liquid scintillation counting and its application to atmospheric ^{85}Kr determination. J Environ Radioact 101(8):615–621
10. Kalinowski MB (1997) Measurements and modelling of atmospheric Krypton-85 as indicator for plutonium separation. International Workshop on the Status of Measurement Techniques for the Identification of Nuclear Signatures, Geel
11. Kalinowski M, Feichter J, Ross O (2006) Atmospheric Krypton-85 transport modeling for verification purposes. INESAP Inf Bull N 27:17–20
12. Takayasu M, Jida T, Watanabe H, Takeishi M, Yamamoto A (2008) Simulation of the atmospheric dispersion of ^{85}Kr from a reprocessing plant over a coastal area. J Radioanal Nucl Chem 275(1):43–54
13. Loosli HH (1984) Haben künstlich erzeugte Raionuclide wie ^{85}Kr, ^{14}C und ^{3}H mit der Luftionisation, mit dem sauren Regen und dem aldsterben etwas zu tun? Separatdruck ans dem SVA Bulletin N3:21–31
14. Harrison RG, ApSimon HM (1994) Krypton-85 pollution and atmospheric electricity. Atmos Environ 28(4):637–648
15. Butkus D, Krenevičius R (1996) Influence of radioactive noble gases on the air ionization variation in the environment of nuclear power plants and nuclear fuel reprocessing plants. Atmos Phys 18(2):43–49
16. Butkus D (1998) Atmospheric ionization caused by ionizing radiation of radioactive noble gas. Aplinkos Inžinerija 6(4):128–132
17. Ulevičius V, Butkus D, Plauškaitė K, Girgždys A, Byčenkienė S, Špirkauskaitė N (2009) Impact of Krypton-85 beta radiation on aerosol particle formation and transformation. Lith J Phys 49(4):471–478
18. Буткус, Донатас. Радиоактивные инертные газы искусствеого происхождения в окружающей среде: научные исследования и течнические решения. Габилитационная работа. Вильнюс [Butkus, D. Radioactive inert gases of the artificial origin in the environment: scientific investigations and technical solutions. Habilitation paper. Vilnius], 1999, 166 c
19. Styra D, Čiučelis A, Usovaitė A, Damauskaitė J (2008) On possibility of short-term prognosis of cyclonic activity aftereffects in Vilnius by variation of hard cosmic ray flux. J Environ Eng Landsc Manag 16(4):159–167
20. Yamamoto T, Tsukui K, Ootsuka N (1984) Storage of Krypton-85 by adsorption method. J Nucl Sci Technol 21(5):372–380
21. Mitev K, Pressyanov D, Dimitrova I, Georgiev S, Boshkova T, Zhivkova V (2009) Measurement of Krypton-85 in water by absorption in polycarbonates. Nucl Instrum Methods Phys Res A 602(3):491–494. ISSN 0168-9002

22. Galamboš M, Kufčakova J, Rajec P (2009) Sorption of stroncium on Slovak betonites. J Radioanal Nucl Chem 281(3):347–357
23. Galamboš M, Paučova V, Kufčakova J, Rosskopfova O, Rajec P, Adamcova R (2010) Cesium sorption on betonites and montmorillonite K10. J Radioanal Nucl Chem 284(1):55–64
24. Gao L, Yang Z, Shi K, Wang X, Guo Z, Wu W (2010) U(VI) sorption on kaolinite: effects of pH, U(VI) concentration and oxyanions. J Radioanal Nucl Chem 284(3):519–526
25. Мартисон ЛК, Малов ЮИ. Диференциальные уравнения математической физики. 2-е издание. Москва: Издательство МГТУ им. Н.Е. Баумана [Martison LK, Malov YuI. Differential equations of mathematical physics], 2002, 368 c. ISBN 5-7038-1911-3