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Journal of Alloys and Compounds 271–273 (1998) 216–220

Journal of ALLOYS AND COMPOUNDS

Up-to-date radioecological situation around the 'Mayak' nuclear facility

B.F. Myasoedov[a,*], E.G. Drozhko[b]

[a]*V.I. Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Acad. Sci., 117975, Kosygin str. 19, Moscow, Russia*
[b]*'Mayak' Production Association, Ministry of Nuclear Energy and Industry of Russia, Moscow, Russia*

Abstract

An overview is given of the modern radioecological situation around the reprocessing plant 'Mayak', which was constructed more than 40 years ago for the production of plutonium for military purposes. The following topics are considered: lake Karachay; artificial water reservoirs contaminated by radionuclides; solid radioactive wastes and their vitrification. Some new approaches, methods and tools developed at the Vernadsky Institute of RAS for determination of different radionuclides in various environmental samples from the impact zone of the facility are discussed. The data on distribution, occurrence forms and migration processes of ^{90}Sr, ^{137}Cs, ^{237}Np, ^{239}Pu and ^{241}Am in aquatic and terrestrial ecosystems are presented. © 1998 Elsevier Science S.A.

Keywords: Radionuclides; Content and speciation in environment; Radiological situation around the plutonium production plant in Russia

1. Introduction

Experimental nuclear tests, nuclear power engineering and numerous accidents that took place at nuclear power plants, atomic submarines and some other nuclear installations made artificial radionuclides a constant and unretrievable component of the modern biosphere, becoming an additional unfavorable ecological factor. The clarifying of the penetration sources, the study of the migration dynamics of radionuclides and the developing of the remediation approaches of the contaminated areas are the most serious ecological, economical and social problems. Solution of these should be based on a wide radioecological monitoring, which demands continual routine analysis of water, soil and various biological samples on contents and occurrence forms of radionuclides.

The present report is devoted to up-to-date radioecological situation around the 'Mayak' nuclear facility and description of some new approaches, methods and tools developed at Vernadsky Institute for determination of different radionuclides in the various environmental samples from impact zone of the facility.

2. The history of the production association 'Mayak'

The production association 'Mayak' (hereafter 'Mayak') is located at 55°44' N and 60°54' E, 70 km north of Chelyabinsk and 15 km east of the town Kyshtym in the southern Urals and covers an area about 200 km^2 including the town Ozersk. 'Mayak' was established in late 40's for production of plutonium for military purposes and for processing fissile materials to achieve a parity in the nuclear arms race. In 1945 a governmental decision was taken on selection of a construction site and the first industrial nuclear reactor was started to be built on the southern shore of the Kysyl-tash, while the settlement for the specialists in the field was built in a peninsula in the southern part of the Irtyash lake [1]. The first continental reactor facility, named 'Annushka', was put into operation on 19 June, 1948. This day may be considered a birthday of 'Mayak' and the nuclear industry of that country as a whole.

Regretfully, at the early stage of running in a number of unique and complicated technologies some serious technical mistakes were made. They aggravated ecological situation around 'Mayak', harmed population and brought about intolerance among local people toward the 'Mayak' activity.

Nowadays, the reactor division of 'Mayak' includes two operating reactors producing radionuclides both for military purpose and general applications and five uranium graphite reactors, which were shut down in 1987–1991. Production of the weapon grade plutonium at 'Mayak' was stopped in 1987. The new plant operation started in 1977 and since then the staff of it has been involved in spent fuel reprocessing from power PWR and FBR reactors of VVER-440, BN-350, BN-600 types, as well as from

*Corresponding author. Tel.: +7 095 1374147; fax: +7 095 9382054.

PII: S0925-8388(98)00057-7

transport and research reactors. For the whole operation period 2380 tons of spent fuel was received from a number of domestic and the foreign power plants. The plant comprises a spent fuel storage pool, three chopping–dissolution process lines, a PUREX extraction with separated plutonium, technetium streams output.

In January 1996 the waste partitioning facility has been put into operation after a prolonged preparatory period. Currently the work is under way on development of the cementation and bituminization facilities for the medium level wastes utilization.

3. Current radioecological situation around 'Mayak'

The region of location of 'Mayak' facilities is a unique area as regards both the scale of environmental contamination and the variety of objects subjected the radiation impact. Significant territories around 'Mayak' have undergone radioactive contamination as result of the 1957 and 1967 accidents and technological discharges into atmosphere. Discharges of liquid wastes to the river Techa at early stages of 'Mayak' operation have resulted in contamination of its floodplane and radiation exposure of the local population inhabiting the shore area.

River 'Techa' belongs to the Iset-Tobol-Irtysh-Ob river system. In its upper reaches it passes 'Mayak'. Liquid waste was released 6 km from the Techa source. About 76 million m^3 of wastes containing 2.7 million Ci of radionuclides were discharged directly into Techa [2]. About 95% of this amount (4300 Ci day^{-1} on the average) was discharged in the period from March 1950 till November 1951 and was deposited within the first 35 km downstream. Most part of these radionuclides has been accumulated in the upper reaches of the river; however, the Iset', Tobol, and Ob' rivers, which successively drain into each other, were also contaminated. The average concentration of ^{90}Sr, ^{137}Cs and ^{239}Pu in the bottom sediments of Techa river in 1993 at the Muslimovo settlement located 78 km downstream amounted to 2.2, 0.3 and 0.025 pCi kg^{-1} respectively.

The accident of 1957 happened due to uncontrolled raise of temperature in the storage tank of the high radioactive wastes of the plutonium production facility which contained large amounts of the sodium nitrite and acetate salts [3]. The liquid sludge ejected to the atmosphere up to the height of 1–2 km formed a radioactive cloud which moved for 345 km to north-east from the storage site and formed a trace on the ground surface due to aerosol precipitation. The region named the Ural Radioactive Trace covered an area of approximately 15 000–23 000 km^2.

The total amount of the radionuclides released to the atmosphere according to different sources varies from 2 to 20 MCi. The major part of radionuclides has been deposited in the vicinity of the accident site. At the closest area 1 to 2 km along the trace axis 0.5–1.0 km wide the density of the soil contamination amounted to 140 000 Ci km^{-2}, while at the area 75 km long and 7 km wide it was equal to 28 Ci km^{-2}.

The dominant part of the radioactive release consisted of the short-lived fission products (^{106}Ru, ^{144}Ce, ^{90}Sr and little amounts of plutonium and uranium.

Fifty years of operation of the plutonium producing enterprise have resulted in local accumulation of large amounts of radioactive wastes (several hundred million Curies, distributed between the low-, middle- and high level liquid wastes and solid wastes.

3.1. Low level radioactive wastes (LLW)

A complex system of the hydrotechnical works comprising a cascade of industrial reservoirs, designed as the LLW storage sites and a chain of the bypass channels was created in the upper reaches of river Techa from 1950 till 1960, to lessen its radiation burden.

Nowadays about 2500 Ci $year^{-1}$ of radioactive materials enter the cascade of reservoirs. As regards of the entire period of 'Mayak' activity the reservoir chain has accepted about 1.8×10^5 Ci of radionuclides, with the prevailing part of this amount accumulated in the V-10 reservoir.

3.2. Middle level radioactive wastes (MLW)

Removal of the MLW is accomplished mainly via their dumping into the open reservoirs V-9 and V-17 (Table 1). The reservoir V-9 (lake Karachay) has accumulated about 2.5 million m^3 of wastes since 1951. It corresponds to 120 MCi of the total radioactivity. Radionuclides are distributed between the mobile bottom sediments (60%), the loamy bed of the lake (35%) and aqueous phase (5%). In the recent years the annual inflow of wastes into the reservoir runs about 20 000 m^3 with specific activity of $1–2\times10^{-2}$ Ci l^{-1} [4].

The first phase of the MLW processing facility has been building since recently to fulfil the requirement of termina-

Table 1
Some characteristics of the V-9 and V-17 reservoirs

Property		Reservoir	
		V-9	V-17
Water area, km^2		0.16	0.17
Volume, Mm^3		0.3	0.3
Concentration in water, Ci l^{-1}	Strontium-90	1.7×10^{-3}	7.0×10^{-6}
	Cesium-137	1.2×10^{-2}	4.0×10^{-6}
	HTO	5.3×10^{-5}	1.0×10^{-4}
	Total α-activity	6.7×10^{-6}	2.0×10^{-9}
	Total β-activity	1.9×10^{-2}	1.1×10^{-5}
Concentration in bottom sediments, Ci l^{-1}	Strontium-90	0.3	0.12
	Cesium-137	1.4	3.3×10^{-2}
Accumulated, Ci	Water	8.4×10^6	4.5×10^3
	Bottom sediments	1.1×10^8	2.0×10^6
	Total	1.2×10^8	2.0×10^6

tion of the wastes dumping into the lake. It comprises the conditioning, evaporation and bituminization installations and a repository for the storage of the bituminization product.

In summer of 1967 a dust from the shore line of the lake containing about 20 TBq ^{90}Sr and ^{137}Cs in the ratio 1:3 was dispersed by the wind over an area of 1800 km^2 and to a distance of 75 km. In late 70's the decision to close and eliminate the lake Karachay water surface area was taken. Application of technology of filling in the lake with the rock ground materials with the hollow-space concrete blocks has made it possible to reduce the water area from 36 hectares to 16 hectares as of the end of 1995. It has allowed to suppress essentially the wind assisted dispersion of radioactive materials as liquid aerosols from the water surface and to reduce a probability of the radioactive dust dispersion from the dried shore line of the lake in case of a strong whirlwind or tornado passing across the lake area. The complete elimination of this threat can be attained, however, only after finishing the water surface closing project.

Another important aspect of the Lake Karachay problem, which needs an urgent solution, is a continual migration of radioactive pollutants into underground water. Under the action of gravity forces industrial solutions drop down to a roof of confining stratum, migrating further along the underground water horizon toward the discharge sites at depths of 40(60)–100 m as result of the gravitational displacement of fresh waters having a lower specific gravity. Concentration of ^{60}Co, ^{90}Sr, ^{106}Ru and ^{137}Cs in the aquiferous horizon is 20–300 times lower than in the V-9 water, while NO_3^- content in a lenticle under the basin is 3.5–5 times higher compared to the lake water [5].

3.3. High-level radioactive wastes (HLW)

As result of accomplishment of the military program about 19 000 m^3 of HLW with the total radioactivity of 135 MCi have been accumulated. Part of the wastes (about 8000 m^3) originated from extraction process is stored in tanks as highly radioactive nitric acid solutions with the total radioactivity of 200 MCi. In addition, every year about 2000–3000 m^3 of HLW is produced as result of nuclear fuel reprocessing from nuclear power plants and marine reactors (totalled to 100 MCi). Some data on quantity of HLW as of March 1, 1995 are shown in Table 2.

Currently produced HLW originated from nuclear fuel reprocessing are subject to evaporation followed by fluxing with phosphoric acid and sodium nitrate and, finally, to vitrification in a furnace of direct electrical heating with the processing capacity up to 500 l h^{-1}. Table 3 shows some data on amount of wastes having been vitrified over various periods at 'Mayak'. The molten mass is casted into special containers of a 200 l volume, which are assembled in groups of three units inside a larger container for the storage in a temporary repository. The rated specific activity of the molten mass is 2500 Ci l^{-1}, while the specific activity of the glass produced amounts to 200–600 Ci l^{-1}.

The wider scale HLW processing can be accomplished only after their special pretreatment involving partition technologies with application of the electric furnace installation with isolated electrodes or microwave heating unit. Currently a new partition facility for the HLW fractionation incorporating an extraction recovery of Cs and Sr by the chlorinated cobalt dicarbolide is developed. The work is under way on optimization of technological scheme of selective isolation of the long-lived alpha-emitters (uranium, neptunium, plutonium) from radioactive effluents, which is based on their extraction by the bidentate neutral organophosphorus compounds.

3.4. Solid radioactive wastes (SRW)

Total amount of SRW generated over the entire period of the 'Mayak' operation is estimated to be equal to 4×10^5 tons. Annual growth of the SRW amount until 1981 amounted to 1×10^4 ton year^{-1}, diminishing gradually in the later years and about $2–2.5\times10^3$ ton year^{-1} of solid wastes were produced in 1995–1996.

All SRW are being disposed into the corresponding solid waste repositories: the ground types repositories are designed for the low and middle level fraction, while the high level SRW are accommodated within the concrete construction with a multiple isolation from the environment.

Investigation of the lateral and ascending migration of ^{137}Cs, ^{90}Sr and plutonium in the vicinity of the SRW repositories shows that concentration of radionuclides in the adjacent ground layers drops sharply down to very low levels as the distance increases, approaching determination limits of radiochemical techniques applied for analysis [6]

Table 2
Amount of high radioactive wastes accumulated at PA 'Mayak'

Type of waste	Amount, m^3	Total activity, MCi
Suspensions	19 000	135
Nitric acid solutions	11 700	249
Vitrified wastes	1974	246

Table 3
Amount of vitrified wastes

Year	Amount, tons	Activity, MCi
1987–1990	162	3.96
1991	178	28.2
1992	563	77.7
1993	448	46.8
1994	407	57.4
1995	216	31.7
Total	1974	245.76

Table 4
Radionuclides concentration in the surrounding grounds as a function of the distance from the waste-ground surface, Bq kg^{-1}

Distance from wastes, cm	Strontium-90	Cesium-137	Plutonium
0.5	2.1×10^{5}	4.8×10^{3}	1.1×10^{3}
1.5	1.8×10^{5}	1.8×10^{3}	1.08×10^{3}
2.5	1.8×10^{4}	1.3×10^{3}	3.1×10^{2}
3.5	9.5×10^{3}	5.9×10^{2}	1.1×10^{1}
4.5	5.5×10^{2}	2.8×10^{2}	1.1×10^{1}
5.5	7.4×10	Trace amounts	Trace amounts

(Table 4). This indicates that the lateral migration of radionuclides from an outer surface of the solid waste body into surrounding soils proceeds rather slowly (less than 10 cm for 15 years). The similar rate of the radionuclides propagation is observed in case of the ascending migration. Significant concentration of radionuclides were detected within the first 10 cm of the ground layer surrounding the medium level solid waste repository. Estimates of an overall propagation distance for the long-lived radionuclides in soil layer show that it will not exceed 1.5 m for ^{90}Sr and ^{137}Cs over a 300 year period (ascending and lateral migration). Hence, one might expect that radionuclides should never reach the repository surface provided that it would be covered by a clay or loamy layer of ground at least 1.5 m thick.

Descending vertical migration can be considered as a superposition of diffusion in a wet ground layer and a convective transfer by a rain water infiltrated the waste layer. It has been shown that for the period of the repositories existence (15–20 years) radionuclides have completely penetrated the aeration ground layer and approached the ground water layer, as illustrated by data from Table 5. It has been noted that contamination level of the ground water samples taken from the borehole in a close proximity to the SRW body is relatively low and it does not exceed a maximum permissible levels established for drinking water.

So, in conclusion to this part of our presentation we must say that a huge amount of radioactive wastes have been accumulated and stored at ‘Mayak’ (about 1 billion Curies), which represent a great ecological hazard.

4. Environmental monitoring at the ‘Mayak’ area

The long term radiation hazard to the contaminated territories arises primarily from the presence of long-lived radionuclides among which the actinide isotopes play a notable role. Decontamination of ground waters, basins, soils and other natural and technogenic objects from these radionuclides is the most important and very difficult problem.

We have performed radiochemical analysis of liquid and solid samples and determined the content and the forms of occurrence of radionuclides. It should be noted that prediction of migration process is a very complex scientific problem even for such a small location owing to diversity of chemical and mineralogical compositions of environmental matrices.

Radionuclide content in rocks and waters has been determined in accordance with the procedure developed at the Vernadsky Institute [7–10]. It makes it possible to determine several radionuclides out of the same sample which saves labor time spent on sample preparation and dissolving. Analysis of soils, rocks and other solid samples includes air drying, disintegrating (milling), sieving through a 1 m siever, igniting at a temperature of approximately 550°C to destroy the organic matter, which takes several days [11]. Radionuclides from the water samples have been concentrated by evaporation (at the Vernadsky Institute) and ultrafiltration (at the central ‘Mayak’ plant laboratory). The last procedure enables to concentrate plutonium out of the water sample of 2 to 50 l in volume. For the determination of radionuclides in the underground

Table 5
Concentration of radionuclides in grounds as a function of the aeration zone depth, Bq kg^{-1}

Distance from wastes, m	Strontium-90	Cesium-137	Plutonium
0	6100	2900	180
1	5800	950	150
2	2000	860	100
3	210	100	6
5	250	2000	3
7	50	30	7
10	180	35	3
12	60	26	9
14	70	47	17
16	240	15	3
17	110	12	3

water samples collected near the Karachay lake 10 to 100 ml of the sample is enough.

To transfer radionuclides from the ashed residue or the water concentrates it is enough to treat the samples with 7–8 M HNO_3 in presence of potassium bromate after their pretreatment by a mixture of HF and H_2SO_4. Further plutonium concentration and radiochemical separation has been carried out on anionite; for americium and curium the complexation sorbent polyarsenazo-n has been used; for ^{90}Sr – the porous copolymer of sterol with divinylbenzole impregnated with 10% dicyclohexyl-18-crown-6 in tetrachloroethane. This method is pretty selective for ^{90}Sr, but needs its careful separation from ^{210}Pb if the latter is present in comparable quantities.

Determination of neptunium is a more complicated procedure as compared with that of plutonium and americium due to the lower concentrations of the former. The method developed at the Vernadsky Institute is based on the neptunium extraction with potassium phosphotungstate during their sorption on the porous teflon membrane impregnated with 0.5 M solution of trioctylammonium nitrate in toluene, which is followed by luminescent determination. $PbMoO_4$ has been used as a crystallophosphor [10].

To determine the chemical yield of plutonium, americium and curium and neptunium, traces of ^{236}Pu, ^{243}Am and ^{239}Np have been inserted into the solid sample before its ignition or in the water sample before its concentration. Detection limits of the radionuclides determination under abovementioned procedures are shown in Table 6.

To predict migration of radionuclides and to develop the remediation approaches, it is necessary to determine not only the content of radionuclides in particular components of biogeocenoses, but the occurrence forms as well.

As for speciation of radionuclides in solid materials, it seemed most appropriate to determine the geochemical forms of mobility [11]. These data are usually obtained by selective leaching. For loamy materials, which mostly accumulate radionuclides, the fraction of relatively mobile forms is maximum for radiostrontium, whereas for plutonium it usually does not exceed 10%. This fact explains the high concentration of ^{90}Sr in groundwater from various sites of the region as compared to plutonium and radiocesium. It is also interesting that the mobility of radiostrontium is higher than that of its natural analogues, calcium and magnesium. Our results show that the migration ability of radionuclides in the ecosystems in question increases in the following series:

$$^{137}Cs < {}^{239-240}Pu < {}^{241}Am < {}^{90}Sr < {}^{60}Co < {}^{237}Np.$$

The data on distribution of radionuclides ^{90}Sr, ^{137}Cs, ^{237}Np, ^{239}Pu and ^{241}Am between components of various ecosystems of Southern Ural region as well as their forms of occurrence show that ^{90}Sr, ^{237}Np and ^{241}Am mostly involved in compounds of fulvic acids, that's why they have a high mobility in the environment. On the contrarily, considerable amounts of ^{137}Cs and Pu have been found in low soluble humic acids bonded primarily with calcium and relatively low mobile hydroxides.

The data on vertical migration of plutonium, radiostrontium and radiocesium in various types of soils have been obtained and corresponding coefficients have been calculated. We consider that one of the most important factors affecting migration resistance of soil media is the content and nature of organic substances. In particular, the clear dependence between diffusional resistance of upper soil layers to plutonium and radiostrontium mass-transfer and content of humus in these layers has been determined.

Table 6
Radionuclides detection limits using described procedures

Radionuclide	Detection limit, Bq g^{-1}
Co-60	1.2×10^{-3}
Sr-90	4.5×10^{-3}
Np-237	2.6×10^{-6}
Pu-239	1.3×10^{-5}
Am-241	2.6×10^{-5}

5. Conclusion

Everything is being done for 'Mayak' to become a waste-free clean enterprise. We should learn from our old mistakes and never make them again in the future.

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