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Distribution of natural radionuclides in some Russian Non-Nuclear industrial technologies

S. Grashchenko, Z.G. Gritchenko, L.V. Shishkunova, A.S. Aloy

SPA " V.G. Khlopin Radium Institute"

Russia

DISTRIBUTION OF NATURAL RADIONUCLIDES IN SOME RUSSIAN NON-NUCLEAR INDUSTRIAL TECHNOLOGIES

Grashchenko S.M., Gritchenko Z. G., Shishkunova L.V., Aloy A.S.
SPA "V.G. Khlopin Radium Institute", St. Petersburg, Russia

ABSTRACT

The distribution of parent and daughter nuclides in intermediate products of industrial processing was studied to estimate the radiation factor and to clarify the feasibility of concentration of natural radionuclides of the uranium and thorium series in several products and wastes. Paper describes the distribution of natural radionuclides in the processes of iodine recovery from stratal waters by air–desorption method and in pretreatment of artesian water for drinking water supply. Also, the distribution of parent and daughter nuclides in intermediate products and wastes manufacturing cycle of titanium production is described. According to the data obtained lead–210 and polonium–210 are transferred to the dust fraction of the ore thermal smelting. The ordinary lead and zinc are also transferred to this fraction. The finest of dust are more enriched, as compared to coarser fractions, with lead and polonium.

INTRODUCTION

Distribution of natural radionuclides of the uranium and thorium radioactive series in non-nuclear industrial technologies is studied at the Khlopin Radium Institute in connection with detecting radioactive contamination by natural radionuclides at the number of industrial plants not involved to the nuclear fuel cycle.

Foundation of the Radium Institute was pioneered in 1922 by V. I. Vernadsky, which was first head of the institute, and was aimed to integrate investigations in the field of radioactivity, regularities in mechanisms of migration of radionuclides and studies of their deposits, and development of procedures for recovery of radium and other radioactive elements from raw materials.

The extensive studies of oil deposit waters were carried out at the Radium Institute in 1927-1933. It was shown that oil waters often contain high concentration of radium. Since oil waters can be considered as probable source of radium, the radioactivity of all oil fields of the Soviet Union was studied [1, 2]. The results of these studies suggested that account must be taken of high concentration of radium in the stages of oil production.

All natural materials contain some amount of natural radioactive impurities. In any technology, these radionuclides arrive both into the resulting products and into the waste products, which can be solid, liquid, or gaseous. Similar to naturally occurring processes, in processing of natural materials, radionuclides can be concentrated or dispersed according to their physicochemical characteristics. When concentration of radionuclides in the resulting products or waste products increases by several orders, ionizing radiation can present a hazard to the staff. When these radionuclides find their way into the environment, they can present a hazard to the population.

To elucidate the causes and conditions of radioactive contamination we determined concentrations of natural radionuclides in the resulting products and waste products of the technologies being studied. The specific activities of radium-226, thorium-228, and radium-228 were determined by precision gamma-spectrometry. The error in determination of radiation-significant nuclides did not exceed 15%. The specific activities of uranium and thorium isotopes and polonium-210 were determined by the alpha-spectrometric isotope-dilution technique with the use of uranium-232, thorium-234, and polonium-208. The error in determination of alpha-radiating nuclides did not exceed 10%. Radium-226 in aqueous samples was determined by the vacuum-emanation technique, and the ratio between radium-226 and radium-228 activities was determined by gamma-spectrometry after concentration of radium isotopes by

coprecipitation with barium sulfate from the samples 40-50 l in volume. Lead-210 in some samples was determined by low-energy gamma-spectrometry [3, 4]. Ordinary lead and zinc in the dust samples were determined by the spectroscopic technique [3].

DISTRIBUTION OF NATURAL RADIONUCLIDES

In hydrochemical processes

In hydrochemical processes, radionuclides are concentrated by coprecipitation and adsorption.

In production of iodine by air-desorption technique

The air-desorption procedure of recovery of iodine from drilling thermal waters includes the following major stages: acidification of the natural mineralized water within a pipeline with sulfuric acid and isolation of elemental iodine, removal of iodine by an air blowing and its adsorption for further refinement, neutralization of spent processing water to pH 7.0-7.5 by controlled addition of aqueous ammonia., setting of wastewater in a special pond for precipitating suspended matter and pumping into underground horizons for maintaining the pressure of underground water [5].

Since brines contain milligramme amounts of strontium and barium their acidification with sulfuric acid results in formation of suspended alkaline-earth metal sulfates, which are deposited in the inner surface of pipelines and get partially into processing reservoir. Accumulation of the precipitates on the surface of equipment deteriorates the process characteristics that requires discharge of the precipitates and cleaning the equipment and pipelines. Discharged precipitates were disposed on the campus of the plant lack of organization since they were assumed to be not hazardous. The rate of equivalent dose (RED) in locations of the sediments was measured with the use of a DRG-01T dosimeter and reached 1.5-1.7 mR/h at the level of 1 m.

Table 1 presents the results of radiochemical and gamma-spectrometric analyses of the alkaline-earth metal sulfate precipitates sampled in cleaning of the equipment and also kept at the plant territory for several years. These results unambiguously show that radioactive contamination is caused by supply of radium isotopes with drilling water and these radionuclides are coming into concentration from the stage of supplying sulfuric acid.

Table 1 Specific activity of natural radionuclides in sulfate precipitates (Bq/g)

Radionuclide	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
U-234, 238	0.002	0.002			
Th-232	<0.02	<0.02			
Th-230	<0.02	<0.02			
Th-228	17.0	7.0	6.7	13.6	5.2
Ra-228	24.7	25.7	26.8	23.4	38.4
Ra-226	23.0	22.0	21.5	23.8	26.3
Po-210	1.3	0.4			
Pb-210	<2.0	<1.5			

The initial drilling waters contain 1.6-2.0 Bq/l of radium-226 and 2.0-2.6 Bq/l of radium-228. Upon acidification with sulfuric acid, radium isotopes are cocrystallized with barium sulfate and strontium

sulfate, are precipitated, and transfer from the liquid phase to the solid. The radium-226 concentration in the wastewater clarified on standing in the processing reservoir is 0.03-0.07 Bq/l. Thus, essentially all radium isotopes arriving to the surface remain at the plant territory and in the processing reservoir. On prolonged exploitation of this technology approximately 5000 tons of these waste products was accumulated. The specific activity of radium isotopes in these waste products corresponds to the specific activity of radium isotopes in uranium-thorium ore with uranium concentration of 0.18% and thorium concentration of 0.6% [6].

In pretreatment of artesian water for drinking water supply

Distribution of natural radionuclides in pretreatment of artesian water for drinking water supply was studied since radioactive contamination was found at one of the water plants in Lebyazh'e (Lomonosov district, Leningrad region). At this water plant operating since 1960s, the Gdov and intermoraine water-bearing horizons are in operation. Since water of the intermoraine horizon contains up to 30 mg/l of ions of bivalent iron, removal of iron is provided. Waters from two horizons are combined, aerated, and chlorinated for iron oxidation. Precipitation and coagulation are carried out with the use of calcium oxide. The resulting hydroxide precipitate is isolated on sandy filter after that purified water arrives to a collector and then is referred to consumers. Hydroxide precipitates accumulated on sandy charge are intermittently discharged with return flow into a collector for hydroxide precipitates and settled processing water.

The RED in machine hall of a water plant reaches 75-110 mR/h depending on degree of radioactive contamination of sandy filter. Analysis of a sandy filter showed that radioactive contamination is mainly caused by radium isotopes (Table 2).

Table 2 Specific activities of radionuclides in the samples of sand and hydroxide precipitate (Bq/g)

Sample	Pb-210	Ra-226	Ra-228	Th-228
Spent sandy charge		5.7	9.0	1.6
The same		7.9	12.3	2.2
The same	0.12	4.4	7.0	1.5
The same		4.3	7.2	1.5
Hydroxide precipitate		14.3	22.2	5.6

Artesian water of Gdov horizon contains 2.0 ± 0.2 Bq/l of radium-226 and 3.2 ± 0.3 Bq/l of radium-228. Radium-226 concentration in the water of intermoraine horizon ranges from 0.04 to 0.13 Bq/l. Radium concentration in drinking water referred to consumers varied from 0.055 to 0.55 Bq/l depending on combination of waters from two horizons.

The radium isotopes arrived with water from artesian water-supply wells are partially coprecipitated with hydroxide precipitates owing to adsorption and remain partially in purified water. In exploitation of water pipeline the precipitates are accumulated in the tubes. The analysis of the precipitates sampled from these tubes showed that specific activity of radium isotopes reaches 20 Bq/g for some samples.

In ore thermal smelting and dust removal

With the aim to study possibility of concentrating natural radionuclides in the stage of smelting of ilmenite concentrates at the Berezniki titanium-magnesium concern, we determined concentrations of natural radionuclides in the initial products, slags, and waste dust.

The processing scheme of titanium production includes ore thermal smelting (OTS) of ilmenite concentrates which alloy with coke in electric furnaces in the coarse of OTS. In this case, titanium rich slags and cast iron are obtained. The process is attended with the dust evolution. Tables 3 and 4 present the results of analyses of the initial raw materials, cast iron, slags, and the dust fraction, which were withdrawn in various stages of dust catching.

Table 3 Specific activities of natural radionuclides in the products of OTS (Bq/kg)

Product	Th-232	Th-230	Th-228	Ra-228	Ra-226	Po-210
Concentrate	240	220	230	250	210	220
The same	240	220	230	240	220	220
Cast iron	<5	<5	<5	<4	<3	2
The same	<8	<6	<9	<4	<3	2
Slag			440	470	300	18
The same			340	400	300	13
The same			440	410	340	10

Table 4 Specific activities of natural radionuclides in the dust products (Bq/kg)

Sample	Ra-226	Ra-228	Th-228	Pb-210	Po-210	Pb(tot),%	Zn,%
Dust caught in cyclone 1st stage	280	320	310		2300	0.24	0.44
The same	260	380	340	2200	2100		
The same	280	410	340	2500	2300		
The same	350	380	380	4000	3800		
2nd stage	196	276	212		9000	0.52	0.9
The same	320		140	9800	9500		
Dust not caught in cyclone	242	240	124		13,500	0.52	0.8
The same	272	343	98		8500	0.62	0.9

As is evident from data listed in Tables 3 and 4, in OTS, radionuclides are practically not transferred to cast iron remaining in the slags and the dust fraction. Lead and polonium isotopes are practically

completely sublimate and transferred to the dust fraction. In this case, the finer fractions of the dust are enriched with these radionuclides to a greater extent than coarser fractions. From ilmenites containing 0.026% of lead and 0.06% of zinc, also ordinary lead and zinc are sublimate and transferred to the dust fraction. In catching and utilization of the dust behind the cyclone, some precautions must be taken to except excess in permissible limits of inhalation supply of radionuclides into organisms of the staff. Similar distribution of radionuclides was noted in the production of tin during the reducing melting of cassiterite [7]. The parent radionuclides are concentrated in slags and lead, bismuth, and polonium isotopes are concentrated in tin. In purification of tin, polonium isotopes follow to bismuth.
The presented examples of radionuclides distribution in non–nuclear industrial technologies show that at some of them one should take into account the possibility of concentration of natural radionuclides at separate stages of technological process and special precautions are required in handling that materials in accordance with sanitation requirements.

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