Geoecological Assessment of Aged Tailings of Loparite Ore Enrichment / Environmental Assessment of Recycling of Loparite Ore Tailings

E A Krasavtseva1,2, A A Goryachev1,2, V V Maksimova1,2

1Laboratory of Nature-Inspired Technologies and Environmental Safety of the Arctic of the Federal Research Centre «Kola Science Centre of the Russian Academy of Sciences», Fersmana 14, 184209 Apatity, Russia
2Institute of North Industrial Ecology Problems of the Federal Research Centre «Kola Science Centre of the Russian Academy of Sciences», Fersmana 14a, 184209 Apatity, Russia

E-mail: vandeleur2012@yandex.ru

Abstract. Geotechnical properties and material composition of aged tailings of loparite ore enrichment have been investigated. The heterogeneity of the material composition and contents of valuable components has been established. The average content of loparite in tailings is 1.0%. The effective specific activity of natural radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K in the studied samples is in the range of 757.23±158.66 Bq/kg. After leaching samples of tailings with distilled water, the concentration of fluorine ions in the water extract significantly exceeded the maximum permissible concentrations for water bodies of commercial fishing importance, as well as for drinking and household use. Phytotesting allowed to determine that the aqueous extracts of enrichment tailings, both the native extract and its dilutions, do not have a significant toxic effect on the growth of higher plants.

1. Introduction

Tailings dumps negatively impact the natural environment by polluting the surface and ground waters, atmospheric air, and soil layers [1]. Pollution occurs mainly due to dusting of the dump surface, as well as migration of elements with water flows [2]. The negative impact is amplified due to the low resistance of the fragile Arctic ecosystems.

Lovozerskiy GOK LLC extracts loparite ores and produces rare earth metal concentrate, as well as niobium and tantalum. The company operates two dumps of loparite ore enrichment tailings. The first dump has been operated for three decades, after which the company commissioned a new dump for tail slurry storage [3].

The volume of accumulated tailings in the first dump is 6.7 million tons [4]. In this regard, this object is a source of potential pollution of adjacent ecosystems. The most urgent problem is the increased background radiation and high content of easily soluble villiomerite (NaF) in the base material.

Furthermore, the accumulated reserves of tailings in the first dump allow to consider them as a source of valuable elements [5]. Obtaining loparite, nepheline and feldspar concentrates appears to be possible. As no ore mining and crushing operation are required, this object has an additional
attractiveness as a deposit. This determines the relevance of studies aimed at determining the material composition, engineering and geological properties, as well as modeling the migration of pollutants.

The first tailings dump decommissioned 35 years ago is considered in this study to assess the potential negative impact on the surrounding ecosystems, as well as to create a base for further research and development of an optimal waste recycling scheme, minimizing environmental damage and generation of additional commercial products.

2. Research subjects and methods
Samples of tailings were taken from the tailings dump surface by the cutting ring method in 2019 and 2020. Since the central part of the dump is covered with water, sampling was conducted only at peripheral areas. The sampling map diagram is shown in Figure 1.

![Image of sampling points on the tailings dump surface.]

Figure 1. Schematic map of sampling points on the tailings dump surface.

Geotechnical characteristics are determined as per GOST 5180-84 "Soils. Methods for laboratory determination of physical characteristics" [6]. Sieve analysis was conducted using a sieve analyzer with a set of sieves with a mesh size, mm: >1, 1-0.5, 0.5-0.25, 0.25-0.1, 0.1-0.05, <0.05. Chemical analysis was performed using inductively coupled plasma mass spectrometry. The measurements were made on ELAN 9000 DRC-e mass spectrometer (Perkin Elmer, USA).

The specific activities of radium-226, thorium-232, potassium-40 in the samples of tailings of Lovozersky GOK LLC were calculated as per GOST 30108-94 "Building materials and products" [7]. Determination of the specific effective activity of natural radionuclides*. The radioecological analysis of the samples was conducted using the method of gamma spectrometry on the Progress gamma spectrometer.
Mineralogical analysis was performed for two dominant fractions in each of the studied samples using a stereo microscope. To determine the mineral composition of the samples, we used the X-ray powder diffraction method on a DRON-2.0 device, Cu-Kα radiation.

To assess the fluorine migration intensity, the processes of its migration with water flows were modeled in laboratory conditions. The samples of tailings were leached with distilled water. The S:L ratio was 1:4, while the interaction time was varied. Fluorine concentration in solutions was determined by the potentiometric method using an ion-selective electrode Alice – 131F.

Phytotesting method was used for the integral assessment of tailings toxicity. The "Phytotest" method [8] was used to investigate the influence of the aqueous extract of tailings, native extract and several dilutions (blank — distilled water) on the growth and development of higher plants. Plants of two classes were chosen as test cultures — monocotyledonous (common oats Avena sativa L.) and dicotyledonous (watercress Lepidium sativum). After the exposure, the length of seedling roots was measured as a test function. The toxic effect was considered proven if the decrease in test function compared to the blank exceeded 20%.

3. Results

The results of geotechnical investigations led to the conclusion that the tailings are composed of a finely dispersed material with a particle size of less than 1 mm.

Sieve analysis revealed the predominance of 0.5-0.25 mm and 0.25-0.1 mm fractions. Their average content was 24.3% and 28.8%, respectively. The dust fraction (<0.05 mm) accounts for about 12.8%, which can cause dusting in dry windy weather. Some samples contained more than 50% of dust particles. According to the lithological classification, the tailings can be classified as fine and medium-grained sands. The variation coefficient values for each fraction contents indicates the heterogeneity of the grain-size composition of the studied samples. For instance, the coefficient values were 0.66 (0.5-0.25 mm) and 0.49 (0.25-0.1 mm) for the dominant fractions.

The selected tailings are characterized by a fairly high moisture content with the average value among the studied samples — 15.3%. The minimum moisture content was 5.4%, and the maximum was 32.3% (Table 1). Comparison of the grain size distribution and moisture content in the samples allowed to conclude that tailings with a grain size >0.1 mm have high drainage properties. The correlation coefficient of these values was negative for all fractions, except for 0.1-0.05 mm and <0.05 mm (0.76 and 0.88, respectively). As the grain-size distribution of tailings is inhomogeneous, heaving processes and ice lens formation in tailings can be observed during seasonal freezing. Thawing of the surface layer, in turn, can lead to dilution and transition of tailings into a fluid state. Consequently, in the event of a dam failure during the spring flood, the upper thawed layers of artificial soils may be washed out.

The density of tailings in the original burial position varied from 1.52 g/cm³ to 1.99 g/cm³, while the average value was 1.69 g/cm³. With the moisture content increase in tailings, the density of the material increase as well. The correlation coefficient for these indicators was 0.69. The porosity coefficient is correlated with the soil density in its natural state and the dry soil density.

**Table 1.** Main statistical parameters of distribution of engineering and geological properties of aged tailings of loparite ore enrichment (for all samples).

|          | 1   | 2   | 3   | 4   | 5   | 6   |
|----------|-----|-----|-----|-----|-----|-----|
| M (x)    | 1.692 | 1.470 | 2.707 | 45.67 | 15.27 | 0.848 |
| Med (x)  | 1.660 | 1.489 | 2.711 | 44.91 | 12.59 | 0.815 |
| S (x)    | 0.125 | 0.083 | 0.065 | 3.493 | 8.044 | 0.120 |
| V (x)    | 0.074 | 0.056 | 0.024 | 0.076 | 0.527 | 0.141 |
| LDVI     | 1.649 | 1.441 | 2.684 | 44.46 | 12.49 | 0.807 |
| PDVI     | 1.735 | 1.498 | 2.729 | 46.88 | 18.06 | 0.889 |
| Min (x)  | 1.518 | 1.303 | 2.531 | 39.46 | 5.374 | 0.652 |
Max (x) & 1.993 & 1.629 & 2.925 & 52.01 & 32.33 & 1.084  \\
1 – soil density in natural occurrence, g/cm³  \\
2 – dry soil density, g/cm³  \\
3 – true density, g/cm³  \\
4 – porosity, %  \\
5 – soil moisture, %  \\
6 – porosity coefficient  \\
M(x) – average  \\
Medx – median  \\
S(x) – standard deviation  \\
V(x) – variation coefficient  \\
LDVI – lower limit of the confidence interval  \\
PDVI – upper limit of the confidence interval  \\
Min(x) – minimum value  \\
Max(x) – maximum value

The mineral composition of tailings is dominated by potassium-sodium feldspars and nepheline. Pyroxene and amphibole are also present among silicates. Loparite (average content is 1.0%) and eudialyte are present in insignificant amounts, but in some samples, their content exceeds 5% by weight. Reflexes of mainly feldspars (microcline and orthoclase), nepheline and aegirine, were detected in the averaged sample by phase-contrast X-ray analysis. Also, peaks of loparite and sodalite were detected as well. The chemical composition of tailings can be expressed as follows, (%): SiO₂ – 48.0, Al₂O₃ – 18.2, Na₂O – 10.6, K₂O – 4.4, Fe₂O₃ – 5.9, Ti – 0.68, MgO – 0.38, P₂O₅ – 0.82, CaO – 1.7, MnO – 0.22. The following rare earth elements have been detected, (%): Ce – 0.31, La – 0.16. The following elements were detected as well: Zr – 0.38, Nb – 0.12. Considering the significant volume of accumulated waste, the presence of loparite in the studied samples of tailings allows to consider the tailings dump as a technogenic deposit of rare earth elements.

The effective specific activity of natural radionuclides in the average sample is in the range of 757.23±158.66 Bq/kg. Industrial waste with effective specific activity of natural radionuclides of up to 1500 Bq/kg can be sent for burial to industrial waste disposal sites without restrictions by the radiation factor.

Modeling of fluorine migration processes showed that the concentration of fluoride ions in solutions after interaction exceeds the MPC for water bodies of commercial fishing importance (0.75 mg/l), as well as for drinking and household use (1.5 mg/l). Fluorine concentration in the solution increased with the interaction duration increase. After one hour of modeling, the concentration was 0.83 mg/l. The interaction duration increase to five hours led to an increase in the concentration of fluoride ions to 3.09 mg/l. Under natural conditions, such an intensive migration of this element can lead to pollution of the surrounding aquatic ecosystems, which have low self-purification capacity due to northern latitudes.

The phytotesting of water extract of the loparite ore enrichment tailings allowed to find that the most sensitive test culture was common oats Avena sativa L. (Figure 2).
Figure 2. Phytotesting after 7 days: 1 — blank, 2 — native extract, 3 — two-fold dilution, 4 — five-fold dilution.

The decrease in the test function — inhibition of seedling root length — was 10.3% (for the native extract). With further dilution, the negative effect decreased naturally, and the hormesis effect was noted for watercress seeds germinated in a diluted aqueous tailings extract.

4. Conclusions
The conducted studies of aged tailings of loparite ore enrichment at Lovozersky GOK LLC allowed to determine the heterogeneity of the material and grain-size compositions. In this regard, certain areas of the tailings dump with a high content of dusty particles have a high probability of dusting in dry windy weather when the tailings surface has low moisture content.

Mineralogical analysis and phase-contrast X-ray method established the presence of loparite in the studied samples, which allows considering the tailings dump as a technogenic deposit of rare earth elements.

The value of the effective specific activity of natural radionuclides allows to classify the tailings as industrial waste, which can be sent to industrial waste disposal sites without restrictions by the radiation factor.

An intensive migration of fluorine into solution was recorded during leaching of tailings samples with distilled water. At the same time, the concentration of fluoride ions in water exceeds the maximum permissible concentration of water bodies of commercial fishing importance, as well as for drinking and household use, which indicates the potential danger of pollution of natural objects near the tailings dump.

Phytotesting allowed to determine that the aqueous extracts of enrichment tailings, both the native extract and its dilutions, do not have a toxic effect on the growth of higher plants.

5. References
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