

# NORM Related Production of Rare Earth Metals in Estonia

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## Abstract

Since 1970's rare earth, Nb and Ta metals and their compounds are produced in the plant, located at Sillamäe, Estonia. In imported mineral ores, which are used as commercial feedstock materials, NORM concentrations (both <sup>238</sup>U and <sup>232</sup>Th decay chain radionuclides) vary greatly, however they are usually high enough to cause exposure to workers and even sometimes concerns to the public. During processing operations radionuclides become mobilized, migrate to dusts, scales and process residues, leading to the enrichment in these materials. This means that the materials used and NORM waste produced as the by-product of processing, require proper management taking account the safety concerns. At workplaces doses to workers from external exposure, from radon/thoron and dusts in the air are or might be significantly higher than the dose limit for a member of the public. For this reason, these production activities are regulated as a radiation practice.

The paper gives an overview of performed studies and assessments on the impact of NORM, including material and waste streams, radiation exposures during pre-processing, chemical processing, generation of NORM waste, waste management and disposal, their environmental impact, etc. A comprehensive discussion on the establishment of the Estonian regulatory framework for NORM and the arising practical problems is also presented.

## 1. Introduction

In last decades multiple studies have identified the radiological concerns requiring regulatory control in specific NORM industries. EU has introduced the regulation of 'work activities' within its Directive 96/29/EURATOM (EC, 1996) and issued the recommendations for the implementation (EC, 1997). Production of niobium and tantalum, as well as of rare earths, belongs to work activities involving both potentially significant exposure of workers at the work-place (EC, 1999) and potential significance with regard to public exposure as a result of wastes and discharges (EC, 2003). The minerals in niobium (Nb) ores, various concentrates, oxides, etc., raw materials contain enhanced levels of NORM, mostly of <sup>238</sup>U, <sup>232</sup>Th and their decay products. Tantalum (Ta) occurs usually in combination with niobium and rare earths.

A major production facility of Nb, Ta and rare earths, Molycorp Silmet AS, is located at Sillamäe, North East Estonia. The large industrial complex produces the above materials (Nb, Ta metals and light rare earth metals as well as their compounds) from various imported ores and by-products.

The present paper discusses the establishment of the Estonian regulatory framework for NORM, the brief history of the facility at Sillamäe, performed studies and assessments on the impact of NORM at the Silmet facility, problems related to the generation of NORM waste, waste management and disposal, their environmental impact.

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**2. Radiation Act and related legislation**

Estonia is a member state of the European Union since 1st May 2004. Thus, the regulations of the Union are in force in Estonia. When necessary, the Estonian regulations have been modified to take into account the EU regulations. The Radiation Act as the principal legal instrument of the radiation protection infrastructure was brought into force in 1997, while a new upgraded version was enforced in 2004 (Radiation Act, 2004). The Act bases on the concepts, principles, terms, and limits laid down in the International Basic Safety Standards (IAEA, 1995) and Directive 96/29/EURATOM (EC, 1996). The basic internationally approved principles, e.g., justification of practices, optimization of protection and safety, limitation of individual doses, adoption of justified and optimized interventions, the primary responsibility of the licensee, and authorization of practices, are explicitly formulated as provisions of the Act. The EU criteria for the exemption of practices from the requirements of the Act are adopted.

The Act sets requirements for identification and regulation of the work activities relevant to NORM. The general radiation safety principles apply also to the management of radioactive waste, including NORM waste, as well as those arising from decommissioning of a nuclear facility. According to the definitions given in the article 3 of the Radiation Act, radioactive waste is any material or object which contains or is contaminated by radionuclides, the activity or activity concentration of which exceeds the established clearance levels and for which no future use is foreseen.

The licensee in radiation practice is required to take any measures to render harmless radioactive wastes arising from its operation. The Regulation of the Minister of Environment No 10 (2005) issued under the Radiation Act specifies the requirements for radioactive waste management. Radioactive waste will be categorized by activity or specific activity, by half-life, by type of radiation and by heat generation as a result of radioactive decay. In conditioning and storing of radioactive waste their producer has to take into account, beside their type, also physical, chemical and biological properties of radioactive waste. Radioactive waste categorization includes NORM waste, which are defined as radioactive waste arising from processing of natural radionuclides, the activity concentration of which is higher than the exemption levels.

Article 59 of the Radiation Act sets that the dispersion, clearance and management of NORM waste, including the way of their storage, interim storage and disposal shall be determined by the license conditions. The Government Regulation No 163 (2004) enforces exemption levels for radionuclide activity and activity concentration in accordance with the terms and levels equal to those stipulated in the BSS (IAEA, 1995) and the EU Directive (EC, 1999, 2003). Exemption levels are considered as basic criteria for decisions on licensing radiation practices. No license is needed for operations with activities or activity concentrations of radionuclides below the exemption levels. Examples of the exemption levels relevant to the raw material and waste containing NORM are given in Table 1.

**Table 1. Exemption levels for NORM radionuclides**

<b>Radionuclide</b>	<b>Activity (Bq)</b>	<b>Activity concentration (kBq/kg)</b>
$^{210}\text{Pb}$ , $^{226}\text{Ra}$ , $^{235}\text{U}$ , $^{238}\text{U}$	$10^4$	10
$^{228}\text{Th}$ , $^{230}\text{Th}$	$10^4$	1
$^{232}\text{Th nat}$ , $^{238}\text{U nat}$	$10^3$	1

For multiple radionuclides or mixtures in the materials, the sum of their activity or specific activity ratios to the corresponding exemption levels should be less than 1.

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Environmental impact assessment procedure is required for radioactive waste management facilities, as they are considered in the legislation as activities with a significant environmental impact (EIA, 2005).

In 2009 in the course of reorganization, the Estonian Radiation Protection Centre, the former authority since 1996, was merged as a department with the Environmental Board. It is empowered to authorize practices by licensing, to assess practices and sources, to maintain the dose and source registers, to monitor and to assess radiation levels, to implement international conventions and agreements, to notify about the radiation accidents, etc. The other body, the Environmental Inspectorate, is provided to carry out regular inspections of the licensed radiation practices.

### 3. History of the Silmet facility

The large industrial complex at Sillamäe, about 190 km East from Tallinn, was launched as a top secret facility in 1948 for mining and milling of local alum shale (*dictyonema argillite*) containing ~ 0.03 % of U. Before Estonia regained independence, the facility under different names, including the Sillamäe Metallurgy Plant, was managed by the former USSR Ministry of Medium-Scale Engineering and it produced uranium for military and civil use. Later the mines were closed and much richer uranium ore of up to 1 % of U was imported from the Eastern European countries. Waste arising from uranium production was stored in a depository located near the Sillamäe plant, 20 - 50 m from the waterline of the Baltic Sea. After processing as a total of about 4 million tons of uranium ore, the uranium production was closed in 1977.

In the beginning of the 1970s the facility was modified for production of niobium, tantalum and rare earth metals, using loparite as a NORM-containing raw mineral from the Kola Peninsula. Later (till now) rare earths were produced from rare earth chloride mix. Composition of raw materials varies depending on the deposit, as niobium/tantalum are usually combined with iron, tin, titanium, manganese, radioactive elements (uranium, thorium) and their decay products. The composition and amount of technological waste from the processing depends on the share of each raw material type in its total amount. As the waste contained small amounts of thorium and uranium as well as their decay products, which were not recovered, the arising NORM waste were dumped together with other waste to the pond on top of uranium tailings depository near the plant. Since 1990 the main activity of the plant has been the continuation of the production of Nb and Ta metals and light rare earth metals as well as their compounds from various imported ores, e.g., columbite and chloride melts. In 1992 the facility becomes the state joint-stock company RAS Silmet, later AS Silmet and now Molycorp Silmet AS, which continues the production of Nb, Ta and rare earths.

With the establishment of the radiation protection infrastructure in Estonia, the NORM related working activities at the Silmet facility were considered of radiological concern, which required regulation as a licensed radiation practice. The performed studies, showed that at workplaces doses to workers from external exposure, from radon/thoron and dusts in the air were or might be higher than the established dose limits for a member of the public (see, e.g., Mustonen, R., et al., 2000). In addition, an analysis showed that the use of the former uranium tailings depository for dumping of the NORM waste might cause some radiological concerns to the members of the public (Realo, 2000).

### 4. Need for new NORM-waste management system

Until 2004 all radioactive waste from the rare earth and the rare metal production was dumped in the tailings pond. The first environmental impact assessment for the tailing pond was done in 1994 (Ehdwall et al, 1994, Nordlinder. S, et al. 1995). Mostly because of the impacts caused by releases and discharges of chemical pollutants from the depository and from the pond on its top, an international PHARE remediation project was initiated and the use of depository was terminated. The remediation project was

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successfully finished in 2008. As a result, the need arose to work out a new waste management option for the produced NORM waste. For creation of the new waste management system according to the Radiation Act and waste laws, international agreements and other legal acts, a number of assessments and studies there have been carried out.

It became clear that future radioactive waste arising would be caused exclusively by the production process of the Silmet facility (CASSIOPEE, 2002). The volume and activity of radioactive waste to store or respectively to dispose of would depend on from the following:

- content of NORM radionuclides in the raw materials,
- amount of processed raw materials,
- waste management system.

In the period 2001-2003 the developed radioactive waste management system included radioactive waste separation at an early stage of the technological process. The volume of future radioactive waste arising was estimated to be maximum 2000 t/y before vitrification. The specific  $\alpha$ -activity of about 7000 Bq/g was estimated for non-vitrified radioactive waste. It was considered that all radioactive waste should be treated and conditioned together and after packaging in the special containers, it would be stored in an interim storage facility of a modular type. After 50-100 years of the interim storage period the waste could be used as feedstock for further processing or disposed of in a final repository (Behre Dolbear & Company, 2002). In case of the final disposal, the waste vitrification option was considered. The vitrified radioactive waste would be dumped into the existing oil-shale ash storage of the local power plant located at the Western side of the former tailings pond dam. This solution represented practically a final near-surface disposal of vitrified long-lived radioactive waste.

## 5. Environmental impact assessment for NORM waste management

The EIA process for all waste management systems of the facility (including also other forms of waste produced at the plant) was initiated in 2001 (E-Konsult, 2003). As the EIA report provided limited information about the proposed management system for the NORM waste and about the proposed guarantees or assessments for financing of the management options, a special EIA process of the NORM waste management was started in 2003 (E-Konsult, 2004). To meet the public concerns, the EIA program was amended and points covering the possibilities for the future waste management were included. This EIA process was finished in June 2004 and the proposed NORM waste management system was approved.

As the starting point of the EIA process it was taken into account that annually up to 2000 t of NORM waste with activity concentrations of 3000-4000 Bq/g were produced. The estimated amounts and activities of waste are given in Table 2. It was planned that this waste in the drums should be stored temporarily outdoors before the interim storage facility would be finished. In the same time it was expected that after 50 year storage period there would be enough material collected to be of interest to the reprocessing companies, e.g., in Russia or elsewhere.

**Table 2. Estimate of NORM-waste in 2003**

<b>Production line</b>	<b>Average amount of waste per 1 t of processed raw material (kg)</b>	<b>Average activity concentrations of waste (Bq/g)</b>	<b>Estimated annual amounts of waste (t)</b>
Rare earth metals	300-350	4300	1400
Rare metals	170-200	2300	600

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Three different storage packages were investigated:

1. concrete containers with the dimensions of 1.63x1.63x1.35 m, which could contain up to 2.1 tons of the solidified NORM-waste. Putting these containers in 4 layers, would allocate up to 3.15 t of waste on 1 m<sup>2</sup> of the storage,
2. containers used in sea transportation, where the waste would be put in the plastic bags,
3. metal drums with the plastic inside cover of the volume of 0.43 m<sup>3</sup> for or 0.38 t of waste.

As the waste contains <sup>235</sup>U and <sup>232</sup>Th with their decay products, including radon, after several assessments and practical experiments the preference was given to the last option. Radon was estimated as the major factor in causing doses for the radiation workers. The studies resulted in the conclusion that the best solution was the use of double package, which should avoid the leakage of radon for at least 10 y.

The first stage of the EIA process resulted in rather high dose estimates to the radiation workers, e.g., with annual doses over 20 mSv at some operations. A significant overestimation of doses, as it appeared later, was mostly due to the fact that at the start only few real data were available and that many default values and extremely conservative assumptions were used in the assessments. After data corrections and considering realistic protective measures, more realistic dose assessments were performed. As a result of these additional improvements the estimated average annual doses to the radiation workers remained under 4 mSv. The maximum annual doses of about 15 mSv/y, requiring limited working hours in that area were identified for the workers at the packaging facility.

In the safety assessment a number of accident scenarios were also considered:

1. falling and breaking of the drum containing the solid NORM waste in the packaging area or during the transportation;
2. falling and breaking of the drum containing the solid NORM waste in the interim storage;
3. fire in the storage of raw material or in the interim storage of NORM waste;
4. release of the material in the production process.

Based on the assessments and changes on the market, the Silmet plant started to import raw materials with significantly lower NORM radionuclide concentrations, which had resulted in the decrease of produced annually waste volumes by more than 10 times. E.g., they managed to find raw material for the rare earth metal production, which contained NORM below the exemption levels. Nevertheless, the production of Nb and Ta still uses radioactive raw material and the NORM waste generation continues.

## 6. Radiation practice license

Based on the radiation practice licence No 08/004, the Silmet plant is allowed to generate annually no more than 48 t of radioactive NORM containing waste with the activity concentration lower than 300 kBq/kg. At the facility, the generation of NORM waste is not constant in time and it depends to a great extent on the specific production line and the ore used. There is more than 31 t of NORM waste with the average <sup>238</sup>U and <sup>232</sup>Th activity concentrations of 98.8 kBq/kg and 36 kBq/kg, respectively, in the temporary storage. The composition and amount of the processing waste depends on the fraction of each raw material type in the total amount and on their Th, U and their progeny composition.

Under the Radiation Act, the producer of radioactive waste should transfer the arising waste to the radioactive waste management operator in at least 5 years. Unfortunately, there is no radioactive waste management operator for NORM waste in Estonia. At the moment, the NORM waste produced by the Silmet plant is temporarily stored and the company is continuing the search for possible management

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solutions in the future. Unfortunately, so far without any success, as the amount of produced NORM waste is too small for further processing, while their activity concentrations significantly vary. One of the most realistic proposed management options might be the clearance of NORM waste under specified conditions.

## 7. Possible clearance option

The Silmet facility has a functioning power plant, which uses local oil-shale for producing of energy. The estimated annual production of oil-shale ash is around 100 000 t. The Estonian Environmental Board has allowed the use of oil-shale ash in the construction of the Sillamäe harbour, which is built close to the remediated waste depository. In the Environmental Impact Assessment of the Sillamäe Harbour it is estimated that the harbour building needs about 8.45 million m<sup>3</sup> of filling material, including about 2-3 million m<sup>3</sup> of oil-shale ash. Taking account the activity concentrations of NORM in oil-shale ash in Table 3 (Realo et al, 1996) and similarity of physical properties of oil-shale ash and the NORM waste produced in Sillamäe, a clearance option for possible management of the latter has been proposed. The clearance option bases on the assumption that the NORM waste and oil-shale mix (110000 tons of oil-shale ash together with 48 tons of NORM-waste) could be used (instead of oil-shale ash only) in the construction of the harbour.

**Table 3. Activity concentrations of radionuclides in the Estonian oil-shale ash**

<b>Radionuclide</b>	<b>Activity concentrations (Bq/kg)</b>
<sup>226</sup> Ra	48 ... 78
<sup>238</sup> U	48 ... 64
<sup>235</sup> U	2.2 ... 3.0
<sup>232</sup> Th	23 ... 30
<sup>40</sup> K	530 ... 1100

The clearance levels of 1 kBq/kg for both <sup>235</sup>U and <sup>232</sup>Th based on the Radiation Act. The legislation also states that clearance of radioactive waste is possible if:

- the caused annual dose to the public is lower than 0.01 mSv;
- the caused collective annual dose is lower than 1 manSv;
- in the case of the NORM-containing material and waste, the public to the public is lower than 0,3 mSv/y.

The assessment of the annual public and collective doses caused by using the oil-shale and NORM waste mix in the construction of the Sillamäe harbour was performed (Lust, 2009). For the assessment of clearance options of NORM waste an assumption that future radioactive waste arising is caused exclusively by the production of the AS Silmet plant was taken into account. The doses were assessed for both workers and the public considering the following scenarios:

- 1) transportation of NORM waste;
- 2) inhalation in the process of NORM waste management;
- 3) ingestion in the process of NORM waste management;
- 4) fire in the waste management facilities;
- 5) doses to the harbour workers;
- 6) dose to the farmer, who lives and farms on the harbour area.

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The main results of the assessment are the following. For the workers the highest doses arise via inhalation pathway. The highest doses for the public are characteristic of the farmer, who lives and produces in the area filled using the above mix. However, even in the case of the farmer the clearance requirement of NORM, i.e., annual doses lower than 0.3 mSv, is fulfilled with the probability of 95%. Taking into account that the harbour is been built next to the remediated radioactive waste tailings depository, it is hard to believe that farming is a very realistic scenario.

**8. Conclusions**

The outcome of active discussions and dialog between the operator and the regulator was the development of the waste management system for NORM waste, which, however, currently covers only the short range activities. Based on the performed assessments it was proved that in case using the NORM waste and oil-shale ash mix in the construction of the Sillamäe harbour, the clearance requirements would be fulfilled. Additionally, it can be easily proved that of the proposed management option is the optimal solution considering the type, radionuclide composition and amount of the radioactive waste. The final solution for the NORM-waste management is still under discussion.

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