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Title: Turning a Problem into a Resource: Remediation and Waste Management at the Sillamae Site, Estonia

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INTRODUCTION AND RECOMMENDATIONS

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1. Introduction

Estonia, a country of about 1.5 million people and 45,200 km^2 land area, inherited a number of environmental liabilities from the Soviet Union. One of the largest of these liabilities is the Sillamäe site in northeastern Estonia. A NATO Advanced Research Workshop, "Turning a Problem into a Resource: Remediation and Waste Management at the Sillamäe Site, Estonia," was held in Tallinn, Estonia, 5-9 October 1998. The purpose of that workshop was to bring together experts to discuss relevant experience in other countries and make recommendations on the future course of remediation and waste management at Sillamäe.

The Sillamäe Metallurgy Plant, which was managed by the USSR Ministry of Medium-Scale Engineering, was built in 1946 to produce uranium metal from local shales. The shales were found to be unsatisfactory for this purpose, and uranium ores were imported from central and east European countries, particularly from Hungary and the former Czechoslovakia. During the 1980s, part of the capacity of the plant was converted to production of rare earth oxides and niobium and tantalum, also from imported ores and concentrates. All uranium production at Sillamäe ended in 1990.

After Estonia's independence in 1991, the plant was privatized and now belongs to the Silmet Group. It produces rare earth oxides and niobium and tantalum. A public-private company, ÖkoSil Ltd., 65% owned by the Silmet Group and 35% by the Estonian government, is charged with managing the cleanup of the site and disposal of currently produced wastes.

Environmental liabilities at Sillamäe include a tailings pond of approximately 330,000 m^2 area and 8 million m^3 volume, containing tailings from the metals processing and oil shale ash from power production. The pond is located on the shore of the Gulf of Finland. The stability of the dike, radon and dust emissions from the pond, and uranium and nitrate leaching from the pond into the Gulf must be addressed to bring the site into compliance with environmental regulations. Wastes from current metals processing are discharged to the pond, but this practice must be ended by the year 2002. New technologies and process improvements will be necessary to meet this goal.

This volume is intended to bring together information describing the current situation at Sillamäe and potential solutions based on experience at similar sites as well as promising new technologies. This information will be useful at other sites where uranium and rare earth ores were processed.

In addition to the history of metals production and its concomitant wastes at Sillamäe, northeastern Estonia and nearby parts of Russia have produced oil shale for both chemicals and power production since the early part of this century. The environmental issues at Sillamäe include large amounts of oil shale ash from power production. Solutions to Sillamäe's problems might be extended to the surrounding areas, and considering the region as a whole may well provide better solutions to the environmental problems.

Sillamäe was a secret city, closed to Estonians until Estonian independence. Its population is predominantly ethnic Russian. Environmental and economic problems will complicate the integration of Sillamäe into Estonia. We have therefore tried to address the technical problems in a broad context so that this volume will be useful to city planners, economists, and others with an interest in these issues.

Today the Sillamäe radioactive tailing pond remediation project has started. The design work is financed by European Commission through the PHARE environmental program, and the German company Wismut is performing both conceptual and design planning and engineering. The total remediation cost is estimated at approximately USD 22.5 million, and the remediation should be complete by the end of 2005. Estonia's largest environmental remediation project, financed by the European Union, Estonia, Finland, Sweden, Norway and Denmark was officially launched when the financial Memorandum of Understanding was signed in Tallinn on 13 October 1999. Work will start early in the year 2000.

Additional changes since the conference that should be noted include the bankruptcy of Kiviter Ltd. with subsequent reorganization. The discussion and recommendations in Jaan Uustalu's paper remain relevant to oil shale activity in Ida-Virumaa. Also, although the collapse of the ruble and subsequent economic difficulties in the Russian Federation have affected the Estonian economy, an IMF report issued in September 1999 says the Estonia appears to be recovering from that economic impact. Discussions in this book continue to be relevant to the current economic situation.

2. Recommendations

Recommendations were developed by four working groups at the workshop. The working groups concurred on the need for planning and prioritization of work to be done in remediation and waste management at the Sillamäe site. The recommendations are summarized below.

2.1 OLD WASTE (TAILINGS POND)

Proposed work includes decisions on remedial actions to be taken with regard to the tailings pond. The decision-making process must take into account options concerning place and methods, identification of all negative and positive factors in various time perspectives, relevance and weighting of different attributes (including economic factors), and assuring an appropriate level of discussion and information sharing among stakeholders. The process should include all relevant stakeholders, in particular, Silmet, governmental authorities, governments of countries bordering the Baltic Sea, and the public.

A procedure to develop a good solution must include the master plan as a basis for planning and identification of technical and regulatory requirements on stability, water and radon tightness, dust prevention, erosion, leakage, and root penetration. A risk evaluation should be conducted concerning events that could jeopardize the tailings, including human and animal intrusion, earthquakes, and extreme weather.

The principle of using natural materials should be followed as much as possible. Natural materials have been used and tested before, are cost-effective, need minimal maintenance (which can become zero after 100 years), and can be stable for a thousand years or more. The goals should be to meet the technical and regulatory requirements so that the restored area becomes an aesthetically pleasing landscape that can be used in an unrestricted manner for recreation and other non-residential uses. The choice of materials should be for a solution that will last for a thousand years or more.

Control and steering for the remediation projects will lie with the Estonian Ministry of Environment, the funders, and advisory boards such as the Sillamäe International Expert Reference Group (SIERG). Management of the site will lie with Silmet and ÖkoSil.

A procedure for quality control of work and results is necessary. Project managers should use the ISO 9001 and ISO 14001 standards.

2.2 NEW WASTE (RESULTING FROM PLANT OPERATIONS AND BUILDING DEMOLITION)

Proposed work includes decisions on management of current wastes, process modifications and additions of waste management processes, as well as decisions on the disposition of unusable buildings and equipment. For the decision-making process, Okosil should conduct a multiatribute utility analysis, including affected populations, to optimize solutions and maximize agreement on the multiple factors involved. Those factors include (1) the potential for recycle to improve process economics and reduce waste; (2) options concerning methods of processing; (3) identification of all negative and positive factors in various time perspectives; (4) relevance and weighting of different attributes (including economic factors); and (5) assuring an appropriate level of discussion and information sharing among stakeholders. The process should include all relevant stakeholders, in particular, Silmet, governmental authorities, governments of countries bordering the Baltic Sea, and the public.

A procedure to obtain a good solution must include a comprehensive plan, mobilization of international expertise to assist the plant operator in planning waste management activities, a strategy plan for development of production and marketing, and choices of plans for waste treatment in both the short (before 2003) and long time perspectives. The comprehensive plan should be completed prior to closure of the tailings pond and should include all buildings and facilities at the site. The plan should address characterization, prioritization, decontamination and decommissioning, hot spots, closure, remediation, management of current wastes, and disposal methods. The time frames that need to be considered are more than a thousand years for radionuclides with half-lives greater than a thousand years and shorter time frames for radionuclides with half-lives less than a thousand years, depending on the amounts that are present. The time frame for non-radioactive hazardous wast must be determined.

Responsibilities and management issues are the same as for the old waste.

2.3 REGULATORY ISSUES

Meeting regulatory requirements will be a significant part of the cleanup and waste management at Sillamäe. The Estonian regulatory structure is developing. The following recommendations should help to avoid problems in meeting regulatory requirements.

Relevant international recommendations and guidelines should be identified. Licenses to manage old waste and radiation practice should be applied for. Practical guidelines should be issued on occupational and environmental protection. The applications for licenses should be accompanied by environmental impact assessments (EIAs) as necessary. Communication about environmental risks and present and future activities should be improved. New protection technologies should be applied as much as possible. These technologies may need EIAs. Cooperation with other countries should be continued. Close cooperation of Estonian authorities in EIA issues will be needed.

Some questions need to be answered. The regulations are in flux; what standards will be required? A partial answer to this question appears to be that EU standards with practical guide to operators are likely to be close to the standards that will be issued. Also, what role will the regulatory enforcement community play?

2.4 DATA NEEDS

For the actions recommended above, data will be needed for decisions and for the execution of those decisions. The working groups generated the lists of data needs that are presented in this section. It is extremely important to keep in mind that acquisition of data is expensive and therefore must be appropriately justified. However, a factor that also must be taken into consideration is the cost of not having the data; for example, the costs that might be added during remediation if the structure of the tailings pond is found to be different than is now believed. Justification must be done on a more detailed basis than was allowed by the time constraints of the workshop. However, conceptual justifications for the types of data listed below were provided.

2.4.1 Site Assessment before Remediation

To characterize the tailings, both radioactive and non-radioactive contaminants must be considered. The composition and chemistry of the tailings should be characterized, including a complete metals analysis to determine if economically valuable metals are present, and the location of the loparite tails. In addition, the effluent from the pond and the air emissions should be characterized.

For waste management, the influent streams to the tailings pond must be characterized, and both radioactive and non-radioactive contaminants must be considered.

To characterize the geotechnical stability of site, information on wave erosion, the dam profile, and the dam stability is needed.

In order to develop this information a sampling plan must be developed. The plan must include where to sample, how deep to sample, and how many must be taken. This plan is then a basis for requirements for instrumentation, facilities, and personnel.

To prepare for the shutdown of the tailings pond, a list of options for eliminating liquid wastes discharging to the pond prior to 2003. Additionally, opportunities to find solid wastes onsite that can be disposed of in the tailings pond should be explored. Pilot studies should be started as soon as possible to evaluate fly ash as a possible component of the tailings pond cover.

The potential for relocation of the material in the tailings pond and additional material identified for disposal should be evaluated. This evaluation would include the potential of the alternative disposal site for newly generated wastes as well; the possible beneficial use of the tailings as a resource; and the requirements for physical removal of the materials. The potential for stabilization in place should also be evaluated. This evaluation would include the probability and cost of effective closure and the options available for containment, including cover options. Both scenarios then need to be compared for both short-term and long-term risk, cost, benefits, and social impacts.

Appropriate experts from Estonia and other countries should assess the new data before remediation. Data collection and assessment should take approximately one year.

Responsibilities and management issues are the same as given above for the old waste.

2.4.2 *Site Monitoring during Remediation*

Site monitoring during remediation will cover the same parameters as the site assessment, with some additions. As work proceeds, additional sampling and testing is likely to be desirable to assure that the materials are as predicted by the assessment. A sampling plan should be developed to give criteria for additional sampling and testing.

Additional parameters to be monitored include occupational exposures to radioactive and hazardous contaminants, the quality of the work, and the environmental impact of the remediation work.

All data should be assessed as it is acquired so that the work can be adjusted if necessary.

2.4.3 *Site Monitoring after Remediation*

Site monitoring after remediation will cover the same parameters as the site assessment, adjusted to the circumstances. Monitoring should be continued for 20 years, with consideration of different times for different parameters.

INDUSTRIAL COMPLEX IN NORTHEAST ESTONIA: TECHNICAL, ECONOMIC AND ENVIRONMENTAL ASPECTS

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Estonia, with an area of 45,200 square kilometres and a population of 1.5 million, is the northernmost and smallest of the Baltic States. Its location in an important geographical location between the West and the East has brought both wealth and trouble to this country. During the Soviet occupation, the economy of Estonia was developed according to all-Union interests, relying on heavy resource and energy use, and operating with low efficiency and high pollution.

With the collapse of the Communist system and re-establishment of independence, the democratization of society, transition to private entrepreneurship, and a free market began again in Estonia. Initially, the transition was accompanied by decreases in production and consumption, and in the general standard of living. The initial difficulties were mostly overcome, and now the situation has stabilized and improvement is evident. Co-operation with other European countries is of high priority: Estonia was accepted into the first round of negotiations for European Union membership, and co-operation with other Baltic Sea countries is strongly developed.

There are many indices of dynamic growth in the Estonian economy: GDP growth was 4% in 1996 and 11% in 1997; during the past four years, exports have increased 2.4 times, more than 75% of production is from private enterprises, there are about 400 enterprises with partial or full foreign ownership, restructuring and privatization of the infrastructure (including energy and transportation) have been almost fully accomplished, foreign investment has reached 3-4 billion Estonian kroons (USD 200-300 million) a year.

The aftermath of the Soviet occupation has been hardest felt in the Northeast of Estonia (Ida-Virumaa county). An enforced, one-sided development of industry, the establishment of a monolithic oil shale complex to supply energy to the Northwest region of Russia, and the establishment of military factories led to unfortunate economic, social, and environmental consequences in this district. Hundreds of thousands of unskilled workers were brought here from Russia, changing the demographic landscape. Of the 75,000 inhabitants in 1934 of what is today Ida-Virumaa county, 80% were Estonian; now the population is three times larger, and only 18% are

Estonian. The physical landscape has changed as well: what was primarily an agricultural area has become the most polluted district in Estonia.

The Ida-Virumaa district covers an area of 3534 square kilometres. It is bordered on the north by the Finnish Gulf of the Baltic Sea, on the south by Lake Peipsi, on the east by the River Narva, which forms the frontier with the Russian Federation. There are seven towns and sixteen civil parishes in the county. More than 80% of the population lives in the towns—the largest cities are Narva, Kohtla-Järve, and Sillamäe. The largest industries in the district are the oil shale mining, processing and energy-producing complex, the chemical industry (in Kohtla-Järve and Sillamäe), textile factories and machinery (in Narva), and timber and fish processing plants. Altogether, this district accounts for 27% of the industrial production of the whole of Estonia. About the same proportion of the nation's industrial workers live in Ida-Virumaa county, mostly in poorly constructed Soviet-era enclaves, which are currently a source of much anxiety. The infrastructure of the county is relatively good (it includes the Tallinn-St. Petersburg railway and motorway, the Jõhvi-Tartu motorway, seaports, digital telephone and GSM networks, and an adequate energy supply). However, the technical level leaves much to be desired.

About 52% of the county is covered by woodland. Although the county as a whole is quite polluted, in some areas the landscape is still mostly untouched and provides an attractive destination for vacationers and tourists.

Ida-Virumaa is primarily known as a center of the oil shale industry. Nearly 1/3 of its physical area is directly or indirectly connected with the production or utilization of oil shale. Oil shale reserves in Estonia amount to about 3.8 billion tons, half of which can be mined in accordance with existing technological, economic and environmental criteria. The industrial layers have an average thickness of 2.5 metres, and shale beds are at a depth of up to 80 metres, the depth increasing in a southerly direction.

Oil shale consists of 30-40% organic matter (kerogen) and 60-70% mineral substance. The sulfur content is 1.5-1.6%. The net calorific value of oil shale varies from 2000 to 2400 kcal/kg, which is on the average about 1/3 that of coal. However, oil shale kerogen is rich in hydrogen and oxygen, and up to 20% can be converted into oil.

Oil shale output increased from 2 million to 31 million tons between 1940 and 1980 and has gradually decreased since then to 14 million tons per year, currently mined in a fifty-fifty proportion of open pits and underground mines. Mining is rather inefficient; the mining equipment is mostly of Soviet origin and needs modernization; the loss of oil shale is as much as 30%. Mining is also related to severe environmental problems: open pit mine areas must be reforested or recultivated; there are cave-ins from exhausted underground mines, cones of depression form and lead to decreased water levels in local lakes as well as deterioration in the ground water quality and supply for local people. Waste dumps from overburden and oil shale concentration plants are strikingly conspicuous in the mining areas. The mines have to pay taxes for damaging the environment and for using the natural resources. Even with these taxes and the increasing cost of oil shale (it now costs about 100 kroons or USD 7 per ton), oil shale will nonetheless remain the cheapest fuel in Estonia for many years.

Most (85%) of the mined oil shale is used as fuel in two power plants near Narva. Together, the plants have a total capacity of 3,100 MW but are outdated and badly in need of investment for reconstruction. The NRG Company from the United States has submitted a proposal to privatize the power plants to invest in their modernization.

The generation of power in these plants, based on the combustion of pulverized oil shale, reached its peak in 1980 (19 TWh) and has decreased two-fold since then. When oil shale is burned, SO_2 is released. More than 80% of the resulting SO_2 is bound before it leaves the plant by fly ash containing CaO, but 20%, currently amounting to 80,000 tons per year, is released to the atmosphere. Ironically, because of the decrease in electricity production, the mandate required by international convention to decrease SO_2 emissions by 50% by 1998 was fulfilled.

Fly ash is removed from the exhaust gases in cyclones and electrostatic precipitators, but the smallest particles (about 80,000 tons per year) are released into the atmosphere. There are several heavy and toxic metals concentrated in this finest fraction of ash. Because of contact with alkaline fly ash, the rainwater and snow in the Ida-Virumaa region acquire weak alkaline properties, in contrast to the situation in the rest of Estonia where acid rains predominate. The greatest part of the oil shale ash (at present about 6 million tons per year) is deposited in enormous ash fields, which are potentially hazardous to the environment. During rainy times, excess alkaline water is released into the Narva River.

Recently the first experimental installation for absorbing SO_2 from exhaust gases was put into use. It appears that low-temperature combustion at 900-950 C with a fluidized bed technology is most suited to oil shale. With such a procedure, sulfur is entirely bound by the ash.

Oil shale is also used as a fuel in some citywide central heating boilers and in cement production. In limited amounts, oil shale ash is used in agriculture (for the neutralization of acid soils) and in the building materials industry (for the production of construction materials and as an admixture to cement).

About two million tons of the oil shale mined is used in the processing plants of AS Kiviter, which produces about 300,000 tons of crude shale oil per year and more than thirty manufactured articles based on shale oil or synthetic materials. These plants are also antiquated and pollute the atmosphere with volatile organic compounds and the water with phenols and other compounds in their waste water, although the amount of pollution is decreasing. The most harmful waste product is semi-coke, which accumulates in waste piles. The future of the oil-shale-based oil and chemical industry will depend on the efficiency and competitiveness of its production with naphtha crude oil and its corresponding chemical products.

Oil shale now constitutes 62% of the primary energy balance of Estonia and almost 100% of its energy production. The power plants operate below capacity because of reductions in electricity exports. At the present time, the daytime price of 1 kWh of electricity for consumers is about 65 Estonian sents (4.5 US cents), remaining the cheapest electricity in the Baltic region. Nevertheless, the price is continually rising, which is why we must expect an increase in the share of alternative fuels, primarily renewable resources and imported natural gas. Additionally, oil shale use contributes to the production of large amounts of greenhouse gases (carbon dioxide). Development of the oil shale complex must take into account the Kyoto protocol.

There are about 15,000 employees engaged in the oil shale complex. Reducing production leads to the dismissal of workers on grounds of redundancy, a source of serious social problems that will need to be resolved.

Among the products of the chemical complex in Ida-Virumaa, ammonia and urea (by Nitrofert Ltd.), and benzoic acid and its derivatives (Velsicol Ltd.) are noteworthy. Nitrofert Ltd. is planning the construction of a methanol plant, based on natural gas imported from Russia. The chemical-metallurgical plant of the Silmet Group also belongs to the Ida-Virumaa industrial complex, but a review of the activities nad problems of this plant is not a subject of this paper. There are more than 2000 small enterprises in Ida-Virumaa that play a growing role in the sustainable development of this district.

The revitalization of existing industry is of central importance to Ida-Virumaa. Several studies and projects have been prepared to help accomplish this goal.

URANIUM PROCESSING AT SILLAMÄE AND DECOMMISSIONING OF THE TAILINGS

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Black shales are found all over the world. The southeastern Baltoscandian plateau contains Cambrian and Early Ordovician black alum shales. Swedish deposits are Late Cambrian and thus the oldest in Baltoscandia. The Estonian alum shales, known as graptollite argillites or, popularly, dictyonema shales, are younger–they are Early Ordovician (Tremadoc). Economically exploitable Estonian dictyonema shales are located in the northern part of the country in an area 250 km long and 15 to 80 km wide (total area about 11,000 km^2). The layer is 20-30 m deep layer on the northern border of the deposit, and 200-300 m on the southern border. The layer is 1-2 m thick in the east and 3-7 m thick in the west. Estimated resources (of the category P_2) amount to 62.3 billion tons. The most promising area lies in northeast Estonia (612 km^2), with estimated resources of 4.1 billion tons.

Processing of uraniferous Estonian dictyonema shale in Sillamäe started after World War II. Earlier the town housed a Swedish oil-shale processing plant, which was totally destroyed during the war. It was rebuilt as a uranium extraction and processing facility (also called Facility No 7, Oil-Shale Processing Plant, Enterprise P.O.B. P-6685, Sillamäe Metallurgical Factory, now Silmet). The plant was intended to produce uranium from the local dictyonema shale. The first batch of Estonian uranium was produced in Narva at a pilot plant (known as Cloth Dyeing Factory) during the winter of 1944/45. The following memories of Prof. Althausen [1] show how extraordinarily important this Estonian uranium was to the Soviet Union at that time.

In spring 1945, a high-level meeting attended by L. Beria, K. Voroshilov, G. Malenkov, I. Malyshev, and other members of the Soviet government, as well as leading scientists, including academician P. Kapitza and M. Althausen, took place in the Kremlin, Moscow. I. Malyshev reported that a technology for uranium production from the Estonian dictyonema shale had been developed in the All-Union Institute of Mineral Raw Materials (Moscow) and tested at the Narva pilot plant. He showed a piece of this shale and a 3-litre container of the uranium concentrate obtained in Narva. K. Voroshilov added that in case of necessity the uranium for the Soviet atomic industry would be produced from this "clay".

Construction of the Sillamäe plant started in 1946, and the first production of uranium at Sillamäe was at the end of 1948. Everything that went on there was a military secret. It was so secret that most of the workers did not know the purpose of the plant in which they were working. The word "uranium" was taboo. Different names were used instead, such as: product A-9, metal, tar, silicon, tin, lead, or aluminum. The names of the processes were changed as well (for example ore was called sand, filtration – separation, and enrichment – moistening), and the chemicals (soda, sulfuric acid, ammonia) were called Products No 1, 2, 3 etc., and these numbers were often changed.

In 1946, mining of dictyonema shale began. The first part of the mine (Mine No 1) began production in 1947 and continued to operate for five years. The underground mine is shown in Fig. 1 (striped area). In 1952, alum shale mining at the Sillamäe underground mine was shut down because better ores were found elsewhere. The total quantity of the dictyonema shale excavated in the period 1948–1952 was 271,575 tons. Uranium production from this shale was 22.5 tons (as 40% concentrate). Later production was based on richer uranium ores and concentrates from Central Asia, Czechoslovakia, Hungary, Romania, Poland, and particularly East Germany. Beginning in 1970, the plant started producing niobium and tantalum from Kola Peninsula loparite ore. Besides niobium and tantalum, this ore also contains abundant rare earth metals, as well as up to 5–6 kg/t of thorium and 0.2 kg/t of uranium. These metals were at first all discarded as processing waste, but later rare earths were produced. Uranium production from imported uranium ores and concentrates was enormous; about 98,681 metric tons of uranium (mostly as U_3O_8) were produced in 1950–1989. In 1982, processing of enriched (2 to 4.4% ^{235}U) uranium fuel and reconditioning into UO_2 was started. Altogether 1354.7 tons of enriched uranium were processed. The production and processing of uranium at the Sillamäe facility is shown in Table 1.

In December 1989, both uranium production lines were shut down, and uranium processing was altogether discontinued at Sillamäe as the result of political developments in the Baltic States. A most curious and up to now totally unexplained circumstance surrounds the well documented official production losses, which are much higher for enriched uranium UO_2 fuel pellets reconditioning relative to U_3O_8 preparation from the chemical concentrate.

From 1945 to 1959, the processing wastes of the Narva pilot plant and the Sillamäe uranium extraction facility were disposed of in the area of the present Sillamäe tailings pond. Construction of the tailings pond began in the mid-fifties when a nearly 1/3-km^2 area was filled with oil shale ash and processing waste. The disposal area is shown in Fig. 2. In May 1959, construction of the surrounding and intermediate dams of the "A" and "B" reservoirs was begun. The dams were built of local sand, gravel, production tailings, limestone scrap, building refuse, and trash. In the course of construction, the embankment of the dams was not tightened. Pumping of waste into reservoir "A" was stopped in 1962 when the absolute height rose to 12.35 m over sea level. Filling of the "B" reservoir continued until spring 1964. Then it was stopped and filling of reservoir "A" was started again. In August 1964 filling of reservoir "B" was continued and in November building of the dam for reservoir "C" was started. Some waste was also dumped in the old dictyonema shale mines, see Fig. 1.

Up to 1996, the height of the dams extended to 24.5 to 25.5 m above sea level. The lower part of the dams consists of sand, gravel and ash layers that are cemented as the result of water infiltration. The upper part consists mainly of gravel, building refuse, and broken stones. Distribution of weak sandy clay and ash in the embankment is very irregular. By now the dam stability reserve has fallen to as low as 20% in some sections

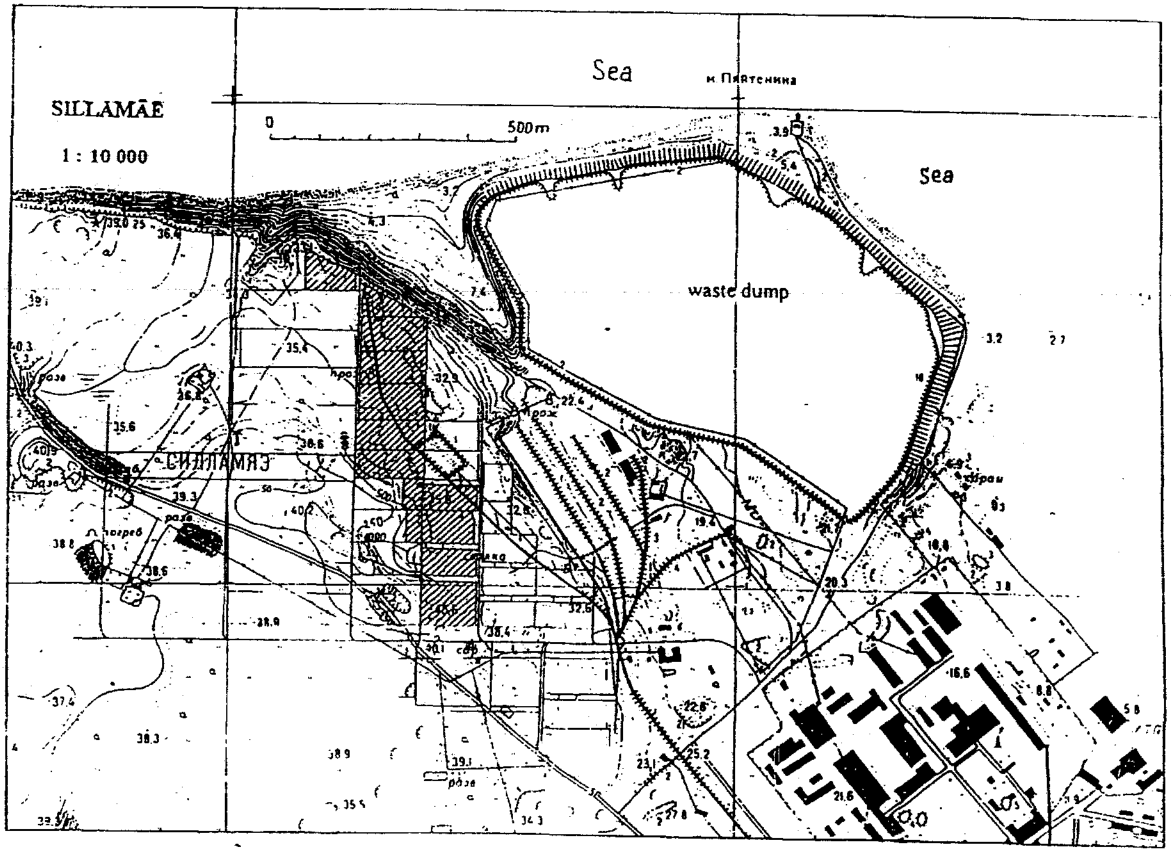

FIGURE 1. Plan View of Sillamäe

TABLE 1. Uranium production at the Sillamäe Metallurgy Plant (also called Industrial Combine No 7, 1946–60; facility No 7, 1961–67; enterprise P.O.B. P-6685) in 1948–1989

Type of Production	Duration	Amount (t)	
		Planned	Actual
Uranium in 40% chemical concentrate, produced from local dictyonema shale	1948-1952	34.7	22.451
Uranium in 40% chemical concentrate, produced from imported ore	1950-1952	187.0	207.0
Uranium in U_3O_8 concentrate, produced from imported ore	1953-1977	23628	24514.5
Total from imported ore	**1950-1977**	**23815**	**24721.5**
Uranium in U_3O_8 concentrate, produced from imported chemical concentrate	1971-1989	73591	73959.3
Total uranium produced from dictyonema shale, imported ore, and imported concentrate	**1948-1989**	**97440.7**	**98703.251**
Uranium in enriched uranium dioxide UO_2 (2 to 4.4% ^{235}U), produced from various enriched raw materials	1982-1989	1476.9	1354.669
Total uranium produced and processed	**1948-1989**		**100054.920**

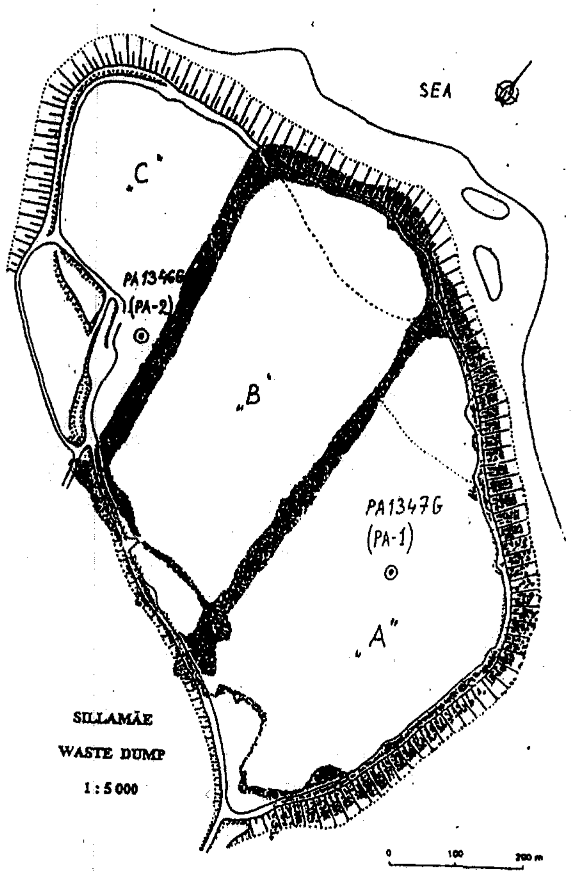

FIGURE 2. Plan View of Disposal Area.

and is thus nearly exhausted. This is why a real and present danger of bursting of the dam exists in case of even a small earthquake or explosion, in which case a large amount of radioactive waste might be swept into the Gulf. Sillamäe is situated in a seismically active area. The last quake of dangerous magnitude took place in nearby Narva in 1881 [2].

In the tailings pond, three depositional layers of waste may be differentiated (Fig. 3):

- a grey layer of sandy clay mixed with ashes /layer 3), consisting of loparite ore processing waste;
- a brown layer of sandy clay (layer 5), consisting of uranium ore processing waste;
- pebbles (layer 6).

For calculating the capacities of the waste layers, a three-dimensional model of the tailings pond was created using the "Terra Modeler" program. In calculating the capacities the "Terra Quantity" program was used. According to this layer capacities expressed in tons are as follows:

• layer 3 ($\delta = 1.4\ t{\cdot}m^{-3}$)	4,000,000 t
• layer 5 ($\delta = 1.55\ t{\cdot}m^{-3}$)	4,052,000 t
• layer 6 ($\delta = 1.7\ t{\cdot}m^{-3}$)	648,000 t

By current estimate, the tailings contain approximately 1200 t of uranium, 800 t of thorium, up to 90-120 kCi of their daughter nuclides, including up to 12 kg of ^{226}Ra. The γ-radiation level on the tailings surface amounts to 100–1700 mkR/h, in places as much as 2000–3000 mkR/h.

The locations of the boreholes PA-1 (1347 G) (depth 20.4 m, ø 89 mm) and PA-2 (1346 G) (depth 16.5 m, ø 89 mm) are shown in Fig. 2. Borehole PA-1 was drilled within the boundaries of the sediment reservoir "A", the filling of which was started in 1959. Borehole PA-2 was drilled within the boundaries of sediment pool "C" which began to be filled at the end of 1964. Gamma-ray logging with the CK-1-74 well logging station was carried out in the boreholes. Only γ-radiation from the waste products surrounding the boreholes was registered. Measuring results for the borehole PA-2 are shown on chart (Fig. 4), expressing the γ-radiation exposure rates (mkR/h) in relation to registration depth. The γ-radiation level amounts to 5000–7000 mkR/h at depth of 5–7 m; in PA-1 7500–9000 mkR/h at 19 m depth.

The floor of the tailings pond lower layer (layer 6, in its absence layer 5) consists of natural gravel and pebbles (0.5–7.9 m thick) and a water-tight layer of Cambrian blue clay underneath. The clay layer has a 0.0025–0.004 surface inclination towards the Gulf.

Hydrogeological conditions in the pond are dominated by close connection between the ground, surface and anthropogenic waters. The composition of the water flowing into the sea from beneath the tailings depends on the same conditions. The waters of the tailings pond drain through the layer of gravel and pebbles into a ditch that empties into the sea. The amount of water flowing daily from the tailings pond into the sea is estimated as 1600 m3. The amount of nitrogen dissolved in water carried into the sea constitutes 1500 tons a year, that of mineral matter – 6000-12000 tons a year. The total mineralization of the water is 10.4 to 21.2 g/l, average about 15 g/l. Earlier studies show that waters from the tailings carry some dissolved natural radionuclides towards the sea, slightly contaminating beach sand and the blue clay layers. The high nitrogen content of

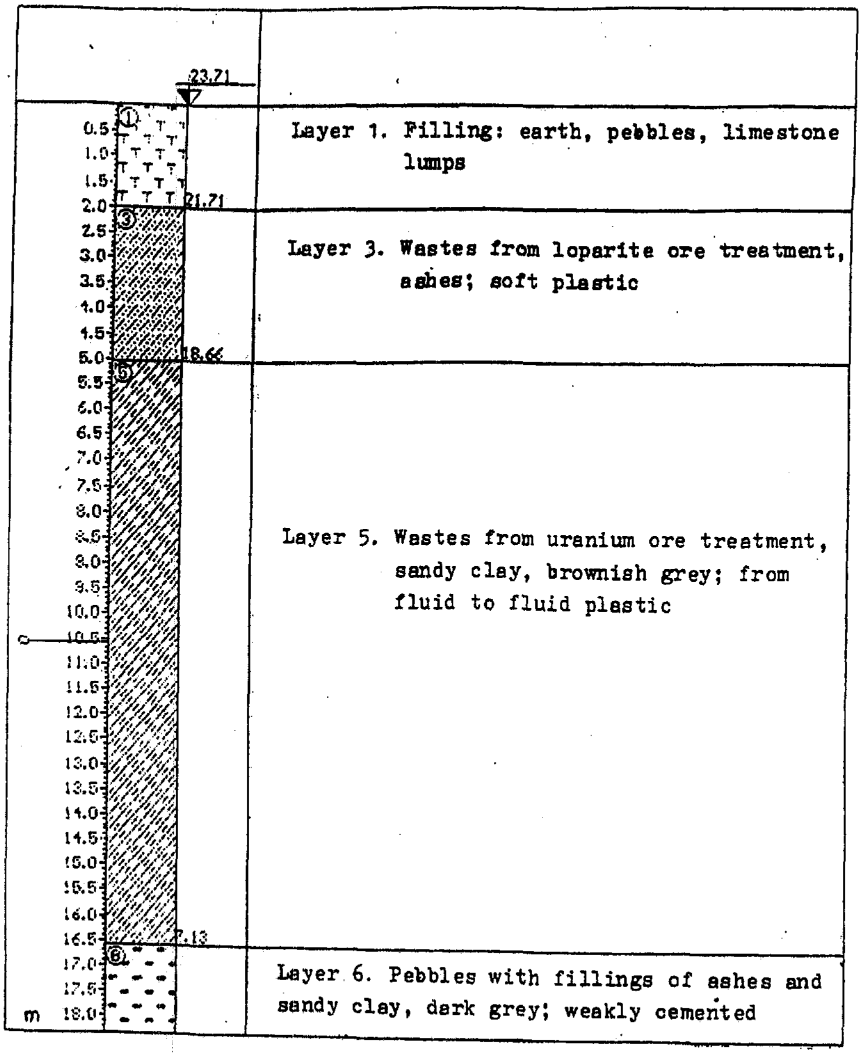

FIGURE 3. Waste layers in the tailings pond.

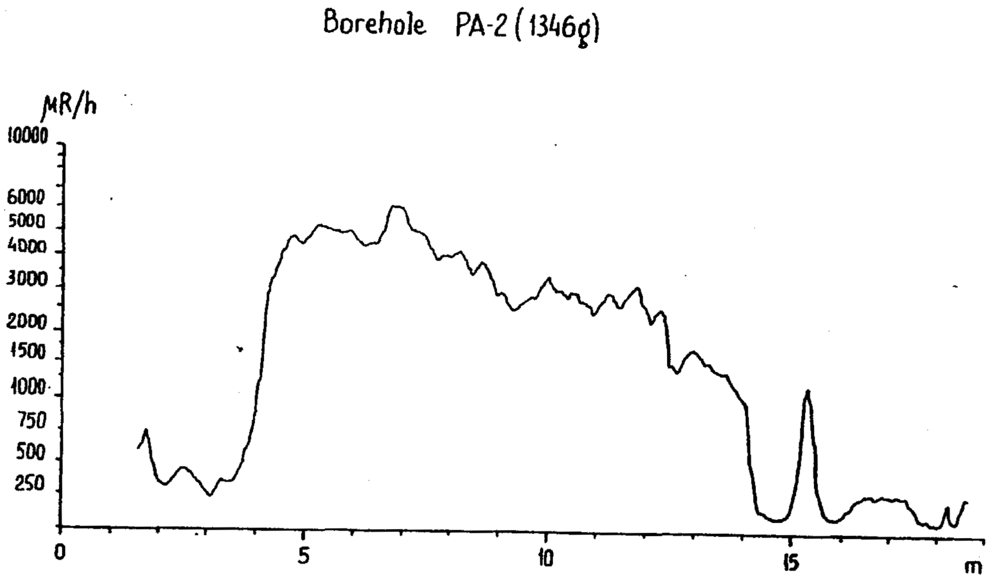

FIGURE 4. Gamma-ray log for Borehole PA-2

the tailings pond water has increased the growth of seaweeds in the coastal water behind the dam.

At Sillamäe, the currents of the sea move predominantly eastward along the coast. This is why the dissolved waste products carried from the tailings pond into the sea or wind-blown towards the town are polluting the town of Sillamäe as well as its eastern beaches. Preliminary data show that the tailings is a real threat to the Gulf of Finland as well as for the town of Sillamäe and its nearby environment for several reasons:

a) About 6 million. m^3 of radioactive (90 to 120 kCi) waste lie behind the dam, the stability reserve of which is practically exhausted. Taking into consideration the flowing consistency and thixotrophy of the deposited material, in case of an explosion or an earthquake the dam would burst and let the processing wastes flow into the sea.

b) The tailings contain about 1200 tons of uranium and a large quantity of various natural radionuclides, providing a direct source of radioactivity. Radionuclides spread into the sea, surface, and ground waters, disperse through the air (wind mobilization of radioactive deposit material and emanation of ^{222}Ra) into the surrounding environment, thus endangering people and the environment in general.

c) Besides radionuclides, the tailings contain numerous other toxic compounds present in the uranium and niobium ores and concentrates, such as arsenic and soluble rare earth salts. There are no data available on the environmental impact of the latter, because these valuable salts are, as a rule, never dumped.

d) The forms of the radioisotopes present, particularly radium isotopes, may become more dangerous with time through biochemical interactions that cause this material to become more soluble.

e) Waters flowing through the tailings and carried out to the sea with ground water continue to transport pollutants into the Gulf. They carry 24 tons of mineral matter leached out of the tailings daily. The high level of nitrogen in tailings water causes eutrophication of the coastal seawater.

In the course of the studies, essential information for estimating the environmental impact of the waste was established. Among the findings:

- Extremely high ^{226}Ra content (12 kg in $RaSO_4$) in the tailings;
- Very uneven distribution of γ-radionuclides in the tailings, but no fission products, no Pu, no α-active or transuranium elements were found;
- Radon can provide difficulties. The half-life of ^{222}Rn is less than 4 days, but it is able to diffuse through at least 5 m of topsoil.

The natural background of radon is about 40 Bq/m^3. In the basements of homes in Sillamäe the measured radon concentration amounted to 10,300 Bq/m^3, exceeding the natural background 250 times and the indoors standard by 70 times [3]. The information given to the government by the state commission founded in 1989 stated that there were places in Sillamäe where the content of radium waste products in the soil exceeded the natural background by 85 times, in the former uranium storehouse more than 2000 times, the content of thorium series waste products in the soil exceeded the background for 73 times, and in the tailings pond up to 160 times.

What could be done with the tailings pond and its surroundings?

The highly polluted area extends about 300 meters from the dam. The polluted soil may be partially removed and the whole area covered with clean soil. The outflow and the accompanying environmental pollution should be prevented or at least limited. The whole surface of the tailings pond could be paved or covered with buried plastic like a refuse dump.

As the ongoing operation of the plant is accompanied by hazardous waste production, these wastes should not be let into the old tailings pond, and possible radioactive elements in the waste should be precipitated as indissoluble substances, melted into glass or concrete blocks and buried in special places as it is done elsewhere in the world. Such a technology is expensive, but unavoidable.

In 1996 the government allocated funds for preliminary reinforcing of the tailings pond. A competition was announced and reinforcement works started. For the whole year the dam was heightened and reinforced from the sea side with granite blocks, concrete, and limestone. These works were carried out by companies from Pärnu and Virumaa county. The estimated cost of proper reinforcement of the dam is, of course, much higher and it is currently being planned under Phare Multi-Country Environmental Programme. The pre-investment planning of the initial remedial measures is described in Report PPEESD-9802 (Chemnitz 30.03.1999) which includes removal of free pond water, interim covering creating a trafficable surface, contouring, final covering of the consolidated tailings surface, and – last but not least – dam stabilization by placing a pile grillage of sufficient depth (15 m) in front of it and penetrating for 1 to 2 m into the stable non-weathered Cambrian clay.

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SILLAMÄE–A COMMON BALTIC CONCERN

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1. Introduction

The Sillamäe plant was, under the Soviet Union, one of the many facilities in the world for production of uranium for the nuclear fuel cycle or nuclear weapons programs. The experience of uranium mining and milling from the point of view of protection of man and the environment has, until recently, been quite discouraging. In many mines there have been high occupational exposures, particularly from radon and its daughter products. Mining has given clear epidemiological evidence of the detrimental health effect on man of occupational radiation exposure.

As regards the environment, the problems or uranium mining and milling are related to the huge amount of waste containing radioactive as well as chemical pollutants. In many places, there has been real leakage of contaminants into the ground water, watercourses, lakes or sea, and airborne releases of dust or gases like radon. Even where releases have not yet occurred, they remain possible. From the 1940s on, the waste from mining and milling, called mill tailings, were not isolated from the environment in a way that corresponds to today's requirements. Therefore remediation programs have started in many countries [1]. However, remediation is difficult and expensive, and other obstacles to action exist. As releases from the other parts of the nuclear fuel cycle become smaller and smaller, the contribution to the global radiological impact of the nuclear fuel cycle in the longer term is dominated by releases from unprotected tailings.

However, tailings can be satisfactorily protected so that the environmental impact is minor in the short term. Unfortunately, the tailings at Sillamäe do not yet belong to that category. Furthermore, the pond is located on the shore of the Gulf of Finland and the Baltic Sea, which makes it a matter of Baltic concern.

In the following, radiological risks are considered, although many principles and conclusions are applicable also to non-radiological risks.

2. What are the Problems with Sillamäe?

The Sillamäe tailings pond, in terms of area, volume, and radionuclide content, is, if not the largest, among the larger tailings depositions in the world [2]. It is 1–2 orders of magnitude larger than the Ranstad tailings in Sweden and is certainly the largest in the Baltic Sea region. The actual and potential risks of the tailings pond are related to uncontrolled releases of radioactive and non-radioactive pollutants to water and to air. The releases contain long-lived isotopes of uranium, radium and thorium, so that the potential radiological risks continue for a very long time–in practice an unlimited time. The question is how far in the future we, our generation, are responsible for the possible environmental consequences. That is a political, economic and ethical question, quite relevant for the Silamäe case.

Other risks are more potential than actual: for instance, a breach of the dam because of erosion, future settlement on the tailings pond, or intentional or unintentional intrusion and use of the tailings material without awareness of the risks.

The methods to reduce these risks are many; for instance, stabilisation of the tailings by chemical means to keep the toxic substances inside the pond and minimise leakage, strengthening the dam to prevent a breach and increase resistance against erosion, building a breakwater against the eroding waves, covering the tailings with ash, moraine or other materials to shield against gamma radiation, reduce leakage of radon gas, and minimise the risk of unintentional intrusion.

The actual releases today in the Sillamäe region of the Baltic Sea from the taililngs pond are relatively small and should not give rise to concern. However, the situation is unstable, which is why efforts to find remediation methods are justified. But these questions should also be answered: Would the consequences of normal or abnormal, controlled or uncontrolled releases to the environment be serious? Would an accidental release caused by a breach of the dam result in high radiation doses? Would settlement or intrusion result in high doses? To comprehend the questions and the answers, it is necessary to refer to the scientific and regulatory development of the protection issues and to the present basis for judgements on protection.

3. Development of Basis for Judgements

Developments in the area of radiation protection over the last 10 years have been based on scientific research in radiation biology and epidemiology, on broader social and ethical aspects, and on greater concern about protection and preservation of the environment. These factors have led to

- decreased limits for the exposure of public and of workers
- deeper concern for protection of future generations and people in other countries from exposures caused by current practices in a country
- increased consideration of the concerns of the public by making information and participation in the decision-making process more available
- increased determination to protect the environment itself from the detrimental effects of radiation on biological diversity and biological resources
- application of the precautionary principle, which implies, *inter alia*, use of concepts like "best available technology" and "best environmental practice."

These developments are reflected in basic international documents, of which the following examples are particularly relevant for Sillamäe.

3.1 THE RIO DECLARATION OF 1992

The Rio Declaration from the UN conference on environment and development announces *inter alia* (not literally quoted)

- States have the responsibility to see that their own activities and practices do not cause harm to the environment of other countries.
- To reach sustainable development, protection of the environment must be an integrated part of the development process and not be considered in isolation.

- States shall co-operate in a spirit of global agreement to preserve, protect and restore the health and integrity of the ecosystem of the earth.
- On a national level, every individual shall have due access to information on environmental issues from public bodies and shall be given an opportunity to take part in the decision-making process.
- In order to protect the environment, the precautionary principle should be applied as far as possible. If there is a threat of serious and irreversible damage to the environment, lack of scientific proof must not be used as an excuse to postpone cost-effective countermeasures to avoid the damage.

3.2 AGENDA 21 FROM THE RIO CONFERENCE 1992

Chapter 22, Safe and environment adjusted management of radioactive waste, from Agenda 21, states that, concerning co-operation and co-ordination at the international and regional levels, states should co-operate with concerned international organisations where appropriate. States shall not promote or permit storage or disposal of high-, medium- or low-level radioactive waste close to the sea environment without having concluded that scientific results, produced according to valid internationally agreed principles and rules, show that such storage or disposal does not lead to unacceptable risks for human beings or the sea environment. Such storage or disposal must not infringe on other legitimate uses of the sea; the precautionary principle should be applied when reasonable.

3.3 IAEA PUBLICATION 1996, RADIATION PROTECTION AND THE SAFETY OF RADIATION SOURCES [3, 4]; INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, ICRP PUBLICATION 60 OF 1990; AND COUNCIL DIRECTIVE 96/29/EURATOM

The standards and directives in IAEA publication 1996 and Council Directive 96/29/Euratom (BSS), respectively, are based on the recommendations by ICRP, Publication 60. The basic recommendations of IRCP and applied in IAEA Safety Series and EU BSS are the following:

1. For proposed and continuing practices,

- No practice involving exposures to radiation should be adopted unless it produces sufficient benefit to the exposed individuals or to society to offset the radiation detriment it causes. (Justification of practice.)
- In relation to any particular source, the magnitude of individual doses, the number of people exposed, and the likelihood of incurring exposures should all be kept as low as reasonably achievable, with economic and social factors taken into account. This procedure should be constrained by restrictions on the doses to individuals (dose constraints), or the risks to individuals in case of potential exposures (risk constraints), so as to limit the inequity that may result from inherent economic and social judgements. (Optimisation of protection.)
- The exposure of individuals resulting from the combination of all the relevant practices should be subject to dose limits, or to some control of risk in the case of potential exposure. These are aimed at ensuring that no individual is exposed to radiation risks that are judged to be unacceptable from these practices in any normal circumstances. Not all sources are susceptible of control by action at the source, and it is necessary to specify the sources to be included as relevant before selecting a dose limit. (Individual dose and risk limits.)

2. For interventions:

- The proposed intervention should do more good than harm, i.e. the reduction in detriment resulting from the reduction in dose should be sufficient to justify the harm and the costs, including social costs, of the intervention.
- The form, scale, and duration of the intervention should be optimised so that the net benefit of the reduction of dose, i.e. the benefit of the reduction in radiation detriment, less the detriment associated with the intervention, should be maximised.

These quoted parts of ICRP Publication 60 are the principal points of the recommendations. An interesting question is the application of these recommendations to the Sillamäe site and the work that is planned there.

In Publication 60 as well as BSS of IAEA and EU, there is a distinction between practice and intervention. Human activities that increase the overall exposure to radiation by introducing new sources, pathways or individuals or modifying the exposure pathways so the exposure increases in terms of number of people or of dose are called practices. Human activities that decrease the overall exposure by removing sources, modifing pathways, or reducing the number of exposed individuals are called interventions.

All publications state that the dose limit for the public does not apply for intervention, but an "action level" should be chosen appropriate to the actual situation. Justification and optimisation apply in all cases. As regards exposure of workers, there are special limits for intervention in emergency situations. However, in a "chronic exposure" situation, for example, exposures to radioactive residues from a past practice that has been terminated, it is stated in BSS/EU that the dose limits for workers are applicable

In the case of the Sillamäe site, it might be concluded that the remediation work would be classified as intervention, while any new waste disposal at the site is a part of a practice. A remediation could be justified on the basis of averted dose caused by normal releases as well as averted potential exposure caused by accelerated leakage, a breach of the dam, settlements, intrusion etc. Included in these considerations should also be averted non-radiological risk, social and public concern, political aspects, international conventions etc.

3.4 JOINT CONVENTION ON THE SAFETY OF SPENT FUEL MANAGEMENT AND ON THE SAFETY OF RADIOACTIVE WASTE MANAGEMENT. GOV/INF/821-GC(41)/INF/12, 22 September 1997. IAEA.

This convention states *inter alia* that (not literally quoted) radioactive waste should, as far as is compatible with the safety of its management, be disposed of in the state in which it was generated.

The Sillamäe tailings were produced under the responsibility of the Soviet Union. That put the situation in legal, international terms in a peculiar light and invites consideration of shared responsibility among concerned countries.

The objectives of the Convention are (not literally quoted)

- To achieve and maintain a high level of safety in radioactive waste management through enhancement of national measures and international co-operation.
- To ensure that there are effective defences against potential hazards, so that individuals, society and the environment are protected from harmful effects of

radiation, now and in the future. The ability of future generations to meet their needs and aspirations shall not be compromised.

- To prevent accidents and to mitigate their consequences.

This is a clear statement of the importance of international co-operation and support and that justified measures must be taken to protect not only current, but also future generations, providing good long term protection.

The Convention shall not apply to the safety of management of spent fuel or radioactive waste within military or defence programmes. However if and when such materials are transferred permanently to and managed within exclusively civilian programs the Convention shall apply.

Some of the waste at Sillamäe may result from a past military program but, as the tailings pond now is under a civil program, the Convention is applicable.

For existing facilities and past practices, the following is said (literally quoted): Each Constraining Party shall in due course take appropriate steps to review:

a) the safety of any radioactive waste management facility existing at the time the Convention enters into force for that Contracting Party and to ensure that, if necessary, all reasonably practicable improvements are made to upgrade the safety of such facility;

b) the results of past practices in order to determine whether any intervention is needed for reasons of radiation protection bearing in mind that the reduction in detriment resulting from the reduction in dose should be sufficient to justify the harm and the costs, including the social costs, of the intervention.

Regarding legislative and regulatory framework, the Convention says that: Each Contracting Party shall establish and maintain a legislative and regulatory framework to govern the safety of radioactive waste management.

3.5 OTHER PUBLICATIONS

There are a great number of other related international documents on the same issues, mainly published by IAEA and OECD/NEA. One of the fundamental problems that are considered in many of these documents is the long-term safety of a repository. It is a relevant problem in connection with high-level waste and long-lived medium- and low-level waste, particularly alpha-radioactive waste. The problem of increasing uncertainty with time is discussed e.g. by IAEA in its publication on safety indicators [5]. If the waste cannot be left without any control, it must be disposed of in a way that makes it safe for a very long time. One method is isolation from accessible environment by the inherent chemical and mechanical safety of the source, by a series of barriers, by distance, and by choice of an unattractive place/environment. Another method is documentation, marking and informing the public. A third method is institutional control. The goal is, of course, to achieve unrestricted use of a site.

All these methods are not mutually exclusive but complementary. All are relevant for uranium mill tailings and for Sillamäe. Because of the long-term perspective needed for protection, the question on reasonable timeframes is raised regarding responsibility, safety guarantees, and uncertainties.

To summarise this complicated discussion including technical, social and ethical aspects it is concluded that:

- quantitative safety assessment of a repository is meaningful for periods up to the order of 1000 years because of increasing uncertainties.

- for longer periods of time, the assessments should be more qualitative than quantitative. For periods longer than 10,000-100,000 years, many aspects of the environment are very uncertain and cannot be predicted.
- institutional control can reasonably be foreseen to be reliable up to a few hundred years.
- the responsibility for safety is on the present generation and the burden (toxic, radiological, institutional etc.) shall be minimal or as low as reasonably achievable. Disruptive events with serious consequences should not occur.

4. Conclusions Relevant to the Sillamäe Tailings Pond

Besides the comments made above related to the various international documents, the following conclusions can be made.

- There are no acute radiation problems today; the risks are long-term.
- The content of radionuclides at the Sillamäe tailings pond is not huge as compared with, for instance, the natural content of same radionuclides in the Baltic Sea. If everything should leak out instantaneously from the tailings pond into the Baltic Sea, it would be easily detected but the resulting doses to people should be very small. The input of alpha radionuclides to the Baltic Sea would be exceptional as compared with releases from other artificial sources like nuclear power plants. There would also be an early increase of activity concentrations in sediments and fish near the Estonian coast and further out.
- The risks of the Sillamäe tailings pond are not considered acceptable in the long term because there are no guarantees of long-term control of risks, and resulting radiation doses from possible scenarios would not be negligible.
- Possible social and economic effects, also in a short-term perspective, of leaving the taililngs pond as it is today should not be underestimated and would also be unacceptable to neighbour countries.
- The Conventions, Declarations, and Standards express heavy international pressure that measures should be taken to improve the situation. Such measures include technical and regulatory matters as well as present and future social and economic aspects and information and participation of the public.

There are many reasons for other countries and international organisations to assist Estonia in this complicated and expensive matter.

Sillamäe is certainly a common Baltic concern.

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MASTER PLAN FOR REMEDIATION OF THE SILLAMÄE TAILINGS POND AND TECHNICAL DESIGN PROJECT

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1. The Master Plan

The Estonian Republic today faces many challenges in the field of environmental protection. The most serious environmental problems are complicated by the fact that they were originated or exacerbated while Estonia was a part of the Soviet Union, and, as a result, responsibilities and liabilities are in question.

Remediation of the Sillamäe radioactive tailings pond is a priority in the Estonian National Environmental Plan [1]. The Sillamäe plant has processed metal ores by hydrometallurgical methods since 1946. Processing started with the production of uranium from local alum shales. Uranium processing continued until 1990, but in the 1970s, production of rare earths and rare metals was introduced and continues today at a smaller scale.

The tailings pond contains the residues from these operations. From 1948 to 1959, waste was disposed of, untreated, at Cape Päite. A dam was constructed there in 1959 to contain the wastes and enlarged at various times after that. Today the tailings pond covers an area of approximately 1000 by 500 m, with a height of approximately 25 m, directly on the shore of the Gulf of Finland (the north-eastern arm of the Baltic Sea).

Briefly, the environmental problems associated with the tailings pond are the stability of the dam and the release of contaminants. The pond was constructed on a slope consisting of several geologic strata. The strata tilt in the direction of the sea. Measurements [2] have shown that the dam is slowly sliding toward the Baltic Sea. Stabilization of the dam is necessary before other remediation schemes, particularly those involving covers, can be applied. In addition, at present, water entering the top of the tailings pond from operations or precipitation moves quickly through the mass of tailings, mobilizing contaminants and carrying them into the Baltic Sea. The flow of contaminants is not in compliance with current standards and regulations, including the Convention of the Baltic Marine Environmental Protection Commission (HELCOM).

In order to deal with these two issues effectively, a master plan was drawn up. The master plan covers the period from 1997 to 2008 and was compiled with the cooperation of the Silmet Group and the Sillamäe International Expert Reference Group (SIERG). The master plan sets up a systematic approach for the overall tailings pond remediation, including drying its interior, reshaping and covering the surface, minimizing water flow through the tailings, and ensuring long-term dam stability.

The master plan consists of five major groups of tasks: remediation of the tailings pond, a future waste management system, long-term monitoring, other decontamination activities, and general planning, enforcement, and surveying. Funding is not complete for all these activities.

The NATO Advanced Research Workshop and this book are a part of the planning process. The recommendations [3] will be incorporated into the master plan.

1.1 TAILINGS POND REMEDIATION

A breakwater was designed and constructed to stop direct wave erosion at the foot of the dam. The planned length of reinforced coastline is approximately 500 m, of which 400 m has been completed.

The project for remediation of the Sillamäe radioactive tailings pond has started. Design work is being financed by the European Commission through the PHARE Environmental program. A German company, Wismut, is performing both conceptual and design planning and engineering. The remediation cost is estimated at about USD 22.5 million, and the project should be complete by the end of 2005. Funding of the project consists of contributions from Estonian Government, the European Union and Baltic Sea neighbor countries–Finland, Sweden, Norway and Denmark. This project is discussed in more detail in Section 2.

1.2 FUTURE WASTE MANAGEMENT SYSTEM

The hydrometallurgical plant operated by Silmet Ltd. currently produces rare earth metals (e.g., cerium, neodymium, and gadolinium) and the rare metals niobium and tantalum. These operations discharge approximately 150,000 m^3 annually of a slurry waste. The objective of the future waste management system is to raise the efficiency of the process and thereby decrease emissions. Waste water utilization and purification technology will be acquired to treat and store the solid wastes and to reuse or treat the waste water.

In addition to the hydrometallurgical plant, the Sillamäe thermal power station produces oil shale ash that has been deposited in the tailings pond. The power station is in need of renovation, and that renovation, including the preferred fuels, will include consideration of waste minimization.

1.3 LONG-TERM MONITORING

Monitoring is being done according to schedules agreed upon with the Estonian Radiation Protection Authority. Financial limitations have led to less geotechnical monitoring than is desirable. Laboratory facilities require equipment upgrades for certification. We expect that the PHARE pilot project will allow for additional monitoring.

1.4 OTHER DECONTAMINATION ACTIVITIES

An area of 10 ha, formerly used for ore storage, has been found to be sufficently above background radiation levels that it will need remediation, possibly including removal of surface soil and disposal in a radioactive waste facility. Additional data collection is needed to determine the extent of this area and the best remediation methods.

Another area of 1.5 ha was reported as being polluted with oil products. This area has been partially investigated.

1.5 GENERAL PLANNING, ENFORCEMENT, AND SURVEYING

This task includes oversight by ÖkoSil Ltd., SIERG, and other oversight bodies in general management, oversight, and advising by international experts.

2. Technical Design Project (PHARE Project)

The European Union, under the PHARE program, has funded a pilot project, "Concept *and design of reshaping and covering the Sillamäe radioactive tailings pond,* particularly in relation to dam stability problems." A contractor for this project has been selected, and, at this writing, design work has begun.

The project has two objectives: (1) to set up a systematic approach for reshaping and covering tailings ponds that considers the consequences for long-term geotechnical dam stability; and (2) to develop a design for covering and reinforcement of the Sillamäe tailings pond.

The project consists of three stages: (1) organizing a systematic method of elaborating options for covering tailings ponds in relation to their effects on dam stability; (2) applying this method to the Sillamäe tailings pond to give a conceptual design and initial cost estimates; and (3) to work out a complete detailed design for the cover system of the tailings pond, including improvements that will ensure adequate long-term stability of the dam and the pond.

2.1 FIRST STAGE

In the first stage of the project, information will be collected on different cover techniques and materials applied in international practice, and conceptual recommendations will be obtained for solving covering and dam stability issues from other central and eastern European countries. This information will be evaluated for suitability for application to the Sillamäe tailings pond. All key parameters will be specified that are required for technical characterization of the existing tailings pond in regard to dam stability, including geotechnical, geological, hydrogeological, and coast processes. Appropriate technical measuring methods will be used where specific data are required. A data base will be developed to record, process, and use the data that have been collected relative to the changes in dam stability at Sillamäe. Criteria for long-term stability will be developed that specify the physical properties and data (for example, water content and pore pressure) that must be met to achieve a stable slope and watertight top of the reshaped tailings pond.

2.2 SECOND STAGE

The second stage will provide the application of the methodology developed in the first stage to the Sillamäe tailings pond and will develop a conceptual design. Available data on the relevant parameters as defined in the first stage will be evaluated and a programme for further field tests and laboratory investigations will be developed. The economic and technical aspects of different cover materials, including cost estimates and local availability, will be compared. Local materials, particularly oil-shale ash from the power station, will be tested for suitability as components of the tailings pond cover. A long-term system will be developed for surveillance of the tailings pond, so that data on key processes will be recorded. The laboratory equipment for monitoring will be improved. This information will then be used to develop a conceptual design for the dam reinforcement and covering of the tailings pond.

2.3 THIRD STAGE

In the third stage, a detailed design for reinforcement of the dam reinforcement and construction of the tailings pond cover will be developed, from which the remediation work can start. The detailed design will include construction technology for the tailings pond cover, a handling system for natural precipitation, a seepage water control system, coastal reinforcement, and reinforcement of the dam to bear the additional load of the cover. Technical drawings for construction work and a budget will be developed. The budget will be based on test results and calculations, including materials, transportation, labor, and other factors.

The second and third stages of the pilot project will constitute the first two parts of the Sillamäe Radioactive Tailings Pond Remediation (SRTPR) Project.

The results of the first of the three stages will be useful in other central and eastern European countries facing dam stability problems connected with tailings pond remediation. Countries other than Estonia facing such issues are Poland, the Czech Republic, Hungary, and Bulgaria.

3 Public-Private Partnership

The hydrometallurgical plant at Sillamäe and its concomitant wastes were part of the nuclear weapons complex of the Soviet Union, a state that has ceased to exist. The Republic of Estonia, a state that never developed any nuclear activities, was faced with handling this plant and its wastes. The hydrometallurgical plant was privatized in 1994 and is now owned by the Silmet Group. To deal with the wastes, the Estonian government established a special company, ÖkoSil Ltd. ÖkoSil is a joint venture–a public-private partnership–between the state and Silmet Group. ÖkoSil is owned 35% by the state and 65% by the Silmet Group.

ÖkoSil is established to implement the following tasks: (1) decommissioning and environmental remediation of the Sillamäe radioactive tailings pond and other environmental impacts on areas owned by the Silmet Group and adjacent properties; (2) provision of environmental, laboratory, and consultant services to Silmet Group and other industrial enterprises; (3) development of cleaner technologies for hydrometallurgy; and (4) development of a hazardous waste processing system, including a hazardous waste incineration and treatment plant, to meet the needs of Estonia and the entire eastern Baltic region.

The larger objective of this public-private partnership is to convert a plant that was part of the Soviet nuclear weapons complex to commercial production in an environmentally sound manner and to eliminate the legacy environmental liabilities while contributing to the economic improvement of the region.

4. References

1. Estonian Ministry of the Environment (1998), *National Environmental Action Plan, Estonia*, Tallinna Raamatutrükikoda, Tallinn.
2. Mets. M., and Torn, H. (1999). Stability of the Dam at Sillamäe Tailings Pond. This volume.
3. Rofer, C. K., and Kaasik, T. (1999). Introduction and Recommendations. This volume.

SITE MONITORING

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1. Introduction

The location of the tailings pond at Sillamäe, its construction, and the nature of its contents require a wide variety of types of monitoring. Ideally, such monitoring would take place at numerous locations around the pond.

The tailings pond is located directly on the coast of the Gulf of Finland, an arm of the Baltic Sea. The distance between the dam foundation and the sea water varies from 10 to 50 meters. The pond was designed and constructed as a filtering facility, to retain solids while allowing liquids to drain into the sea. Thus, a drainage system was constructed under the dam. About 3-5 meters below the drainage system and foundation is a thick layer of waterproof Cambrian clay. This layer is inclined toward the sea. The tailings contain several radionuclides and their daughter products as well as toxic non-radioactive metals and nitrates. Radon is produced by the radionuclides. The dam surrounding the pond and part of the surface of the tailings are covered with vegetation, and the remainder of the tailings surface is covered with water.

A comprehensive monitoring program should address the stability of the structure relative to erosional processes, the composition of gaseous and liquid effluents, the composition of continuing solid and liquid inputs to the pond and their effects on the changing tailings surface, and monitoring of transported contaminants in the surrounding Gulf of Finland and adjacent land areas.

Specifically, such monitoring would include

- Coastline monitoring for wave erosion
- Geotechnical monitoring of the stability of the dam
- Monitoring of the volume and composition of the incoming liquid and solid waste
- Monitoring of the distribution of solid sediment and water distribution on the tailings surface
- Monitoring of drainage water composition
- Monitoring of the adjoining area of the Gulf of Finland.
- Air monitoring
- Radiation monitoring.

Monitoring that has been done or is in progress is described in this paper and observations are summarized.

2. Coastline and Geotechnical Monitoring

During the last decades, intense erosion of the coastline near the tailings pond has been observed. A 300-meter stretch of coastline was washed away, and the water approached the dam foundation as closely as 5 meters. In 1997, emergency repairs were performed on this stretch.

Geotechnical monitoring is done by GIB Ltd. (Estonia) and the Norwegian Geotechnical Institute. The results of geotechnical monitoring are reported in the paper by M. Mets in this book [1].

3. Monitoring of Incoming Waste

Both liquid and solid waste from current Silmet production is sent to the tailings pond. Water is regularly sampled for routine analysis. Aggregated samples are collected for 10-day and 1-month periods and chemically analyzed. The results are then used to calculate the amounts of components entering the pond. More detailed information on these components is given in the paper by V. Petrenko and A. Siinmaa in this book [2].

4. Monitoring of Solid Sediment and Water Distribution on the Tailings Surface

This type of monitoring is performed daily in a visual inspection in order to prevent erosion of the inner surface of the dam. Urgent actions are taken when the threat of erosion appears. Consequently, there has never been an occasion of dam erosion during the existence of the dam.

5. Monitoring of Drainage Water Composition

Because the underlying layer of Cambrian clay is inclined toward the sea, drainage from the tailings pond is in this direction. Eight wells were constructed between the dam and the waterline for sampling of drainage water. Twenty-four parameters are analyzed quarterly, and three elements are analyzed once a year.

The content of seven elements (As, Cd, Cr, Co, Nb, Ni, and Pb) in the drainage water is below the sensitivity of the analysis. The pH level varies from 7.8 to 9.2. The most prevalent components are given in Table 1.

TABLE 1. Most Prevalent Components of Drainage Water

Component	Concentration Range (g/l)
NH_4	1.2 – 5.0
NO_3	0.1 – 0.5
SO_4	5 – 12
Cl	1.3 - 3

The volume of drainage water has decreased in recent years, resulting in a decrease in the absolute level of the water on the tailings surface. In other words, the drainage through the tailings exceeds the total input of drainage water and rain water. During the last observation period, the largest possible amount of water drained from the pond was estimated at 750-800 cubic meters per day.

6. Monitoring of the Adjoining Area of the Baltic Sea

Designated sampling points are located in the Gulf of Finland immediately outside the tailings pond, and at distances of 1, 2, 3, and 5 kilometers away. Some of these points correspond to recreation areas in the town of Sillamäe. Sampling takes place once a month. Chemical analysis of the most important contaminants (NH_4, NO_3, SO_4, and Cl) is performed once a month. Analysis of the other elements is performed quarterly. Table 2 gives the amounts of contaminants observed in sea water near the pond.

TABLE 2. Contaminants Observed in Sea Water

Component	Concentration Range (mg/l)
NH_4	100 - 500
NO_3	20 - 40
SO_4	900 - 1400

At the 1-km sampling point, only ammonia is observed in excess (up to 1-2 mg/l). At the 2-km sampling point, excess amounts of ammonia are also observed (up to 1 mg/l). No contaminants are observed in excess amounts beyond 2 km.

A slightly increased amount of uranium is observed in the sea water (up to 0.2 mg/l) in the immediate vicinity of the pond. The amount of uranium becomes normal at a distance of 300-400 meters.

7. Air Monitoring

Since acidity of the water inside the tailings pond is below neutral (pH 2-7), no emission of chemical contaminants is likely. However, emission of radon (^{223}Rn) is possible. Because of a lack of equipment, radon has not been monitored until recently. Measurements conducted by an international team of experts in 1992-1994 determined radon concentrations in the lower air layer over the pond at 300 Bq/m^3, and at 30 Bq/m^3 in the town of Sillamäe.

8. Radiation Monitoring

Continuing observation of γ-radiation levels at the pond show that the radiation level has stabilized in the range of 3 to 35 μSv/h. This wide range was developed historically and is not changing now. With distance from the pond, γ-radiation levels decrease and become normal at a distance of 100-300 meters.

9. References

1. Mets, M., and Torn, H., Stability of the Dam at the Sillamäe Tailings Pond. This volume.
2. Petrenko, V., and Siinmaa, A., Process Flow Sheet. This volume.

STABILITY OF THE DAM AT THE SILLAMÄE TAILINGS POND

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1. Introduction

The tailings pond at Sillamäe is contained by a dam located 30 to 50 meters from the waters of the Baltic Sea, which provide intense erosional pressure. The tailings pond was begun in the 1950s, at which time the height of the dam was 12 m. It has been increased since then to 25 m. The apparently relatively unplanned buildup of the dam and a lack of maintenance have led to a decrease in the stability of the dam. In the worst case, the dam could fail and release the chemically and radioactively hazardous tailings into the sea. Estonian, Norwegian, Swedish, and Finnish specialists have investigated various aspects of the dam's stability. This paper summarizes their findings and makes recommendations for further work.

2. Construction of the Tailings Pond and Investigations of Dam Stability

2.1 CONSTRUCTION OF THE TAILINGS POND

Material from ore processing at the Soviet Sillamäe Metallurgy Plant was initially deposited directly on the ground at Cape Päite, the location of the current tailings pond, with no confining structure. Some of the material filled in natural depressions and pits. In the summer of 1959, construction started on a dam to contain the material, with the boundaries of the current tailings pond (Fig. 1). The dam was constructed from local gravel. Cross dams formed three compartments. Initially, tailings were sent to the western compartment. Oil shale ash was transported in a water slurry from the power plant and deposited in the middle and eastern compartments. The water was removed through wells. Soil was not compressed in the foundations of the dam.

At the end of 1962, transport of tailings to the western compartment was stopped. The absolute height of the tailings at the pipe mouth was 12.35 m. In 1964, tailings were sent to the middle compartment, and the dam height was raised to 16 m. Tailings were diverted back to the western compartment in the summer of 1964, so that the

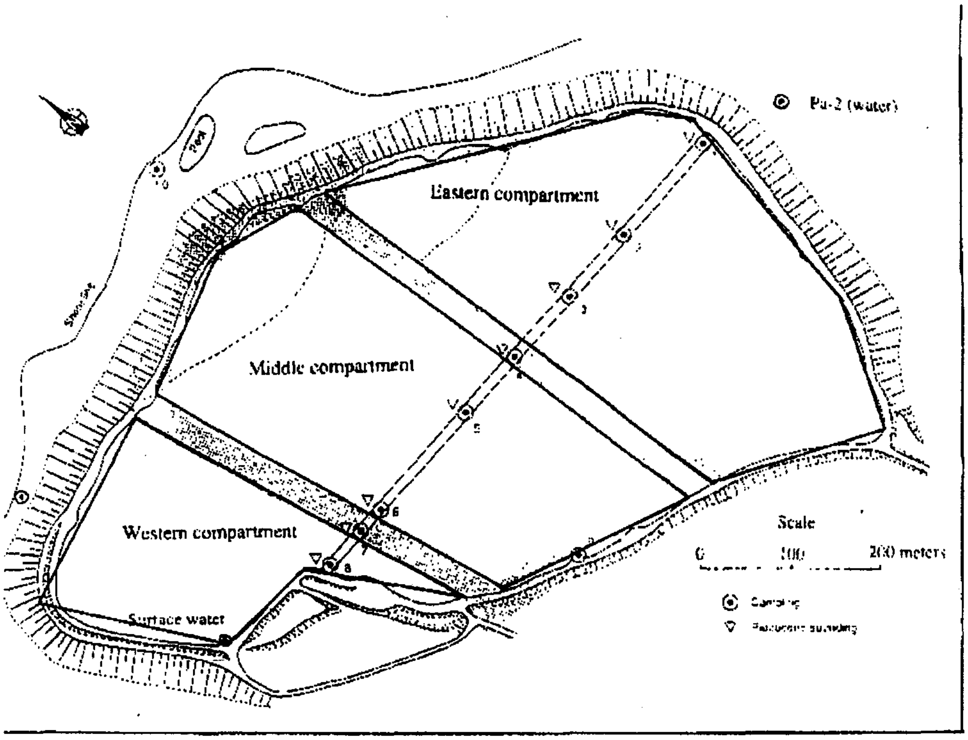

FIGURE 1. Map of the tailings pond.

middle compartment could dry out, and tailings were delivered to the middle compartment after August 1964. At the end of November, construction of the dam to the eastern compartment began.

Records are not available for the period from 1964 to 1984. In 1984, all three compartments were filled, and the dam was raised to an absolute height of 22 m. The current absolute height of the dam is 24.5-25.5 m. The fill was gravel, pit sand, oil shale ash, limestone, debris, and trash, which was not compressed during construction.

Initially, uranium was produced in the Sillamäe Metallurgy Plant from local dictyonema shale. Later, concentrates were imported from Czechoslovakia, Hungary, the German Democratic Republic, Poland, and Romania. Early in the 1970s, loparite processing was started with material from the Kola Peninsula. This material contained uranium and thorium, which were sent to the tailings pond. The plant currently is operating at about 20% of capacity, producing rare earths and rare metals from concentrates imported from Solikamsk.

2.2 SUMMARY OF INVESTIGATIONS 1965 - 1992

The archives of Silmet Ltd., from the Sillamäe Metallurgy Plant, indicate that twelve investigations were made from 1965 to 1992. Four of these investigations [1, 4, 7, 12] are geodetic measurements, six reports [2, 3, 5, 6, 8, 11] describe construction and hydrogeological investigations, and two studies [9, 10] were made to determine the boundaries and properties of soil layers and hydrogeological conditions by geophysical methods.

The geodetic measurements of 1979 [4] and 1992 [12] are the basis for today's mapping efforts. Engineering geological investigations cover the entire area of the tailings pond. They encompass drillings and soil sampling for laboratory tests. The number of field tests has been more limited, although some useful tests on the percolation properties of the soils were done [2, 3, 6, 8, 11].

Hydrogeological investigations have determined the location of watertight and water-conducting strata and their percolation properties, as well as the chemical composition of the water contained [5, 11]. Geophysical investigations have determined the properties of the limestone shore contiguous to the tailings pond, including the weathering crust and the presence of fractures and karst [9, 10].

2.3 SUMMARY OF INVESTIGATIONS 1992 - 1996

Since 1992, the tailings pond at Sillamäe has been investigated almost continuously under a number of different projects. Nine reports have been published [13-21]. In this paper, the existing information on the geological structure of the area and the structure of the tailings pond has been compiled. Additional geotechnical investigations have been made, and a geotechnical model of the area has been developed. A network of geotechnical surveillance points has been established, from which geodynamic processes can be analyzed and the effectiveness of projected stabilizing measures can be evaluated [16-18].

3. Geological Structure and Hydrogeological Conditions

3.1 RELIEF

The tailings pond lies on the pre-limestone shore marine plain, on the first sea terrace, where the absolute heights of the ground fluctuate from 2.2 to 5.5 m. To the west and southwest, the pond borders a Silurian plateau that is surrounded by the limestone shore terrace to the west and north. The surface of the plateau is even, inclining slightly to the southeast.

3.2 GEOLOGICAL STRUCTURE AT THE TAILINGS POND

The dam is built on the first marine terrace, which is composed of marine sediments (m_{IV}). These sediments consist of pebbles or gravel sand for the most part, less often of medium-dense, brown, water-saturated fine or medium sand mixed with ash in the vicinity of the dam. This layer is 0.5-7.9 m thick. The floor of a seam of the layer is the blue clay of the Lower Cambrian Lontova complex.

Eluvial soils (e_{IV}) up to 1.2 m thick lie intermittently on the plateau limestones. They are composed of silty sand and silts.

Deluvial soils (d_{IV}) are distributed within the boundaries of a wide limestone bank belt near the limestone bank. The layer, which is 3.8 m thich, consists mainly of pieces and lumps of silt, sand, and limestone of plastic consistency.

Limnoglacial sediments (lg_{IV}) occur intermittently. They consist of grey, medium dense, water-saturated fine sands or dark grey, plastic silts. The thickness of the sand layer is 0.5 m, and of the silt layer, 10.4 m.

Glacial sediments (g_{III}) occur in the eastern part of the tailings pond and in the region of the Gulf of Sillamäe as grey, plastic silts. Coarse debris makes up 25% of these sediments, and the thickness of the stratum increases to 15 m toward the east.

The bedrock sediments of the area are Lower Ordovician limestones and sandstones and Lower Cambrian clays.

The Lower Ordovician (O_1) occurs on the limestone banks. Sections include limestone of Volhov complex (O_1VI), sandstones of the Mäeküla layer of the Latorp stratum (O_1mk), and limestones of the Pakerord stratum (O_1pk). The limestone of the Volhov stratum (1.7 to 4.3 m thick) lies monoclinally with a slight inclination to the south. The limestone is light grey and dolomitized and contains single pieces of thin marl layers and clay layers. The Mäeküla layer of the Latorp stratum (0.8 to 2.3 m thick) is a greenish-grey, hard, medium or fine grained sandstone, with many grains of glauconite. The argillite (dictyonema limestone shale) layer of the Pakerord stratum is black, hard, and contains greyish-yellow intermediate layers of quartz sand. Its thickness is 0.9 to 3.4 m.

Below the dictyonema layer is a 15.9-m-thick, poorly investigated mixed to finely granular sandstone of Cambrian-Ordovician age (Cm_2-O_1).

Lower Cambrian (Cm_1) Lontova blue clays underlie the entire area, under sediments of Cambrian-Ordovician sandstone or Quaternary sediments. The surface of the blue clays inclines toward the north and lies at the absolute height of 0 to 6 m. The surface of the blue clay is decomposed, and the entire layer contains microfissures. The total thickness of the Cambrian clays is more than 50 m.

3.3 GEOLOGICAL STRUCTURE IN THE IMMEDIATE VICINITY OF THE TAILINGS POND

Natural soils in a section through the tailings pond are marine sediments (m_{IV}), glacial sediments (g_{III}) and Lower Cambrian clays (Cm_1). The anthropogenic soils (t_{IV}) are divided into three groups: gravel-sands, ash, and loams-clays. Their properties are very variable because of their different origins and transport methods. The geological structure of the pond is shown in Fig. 2.

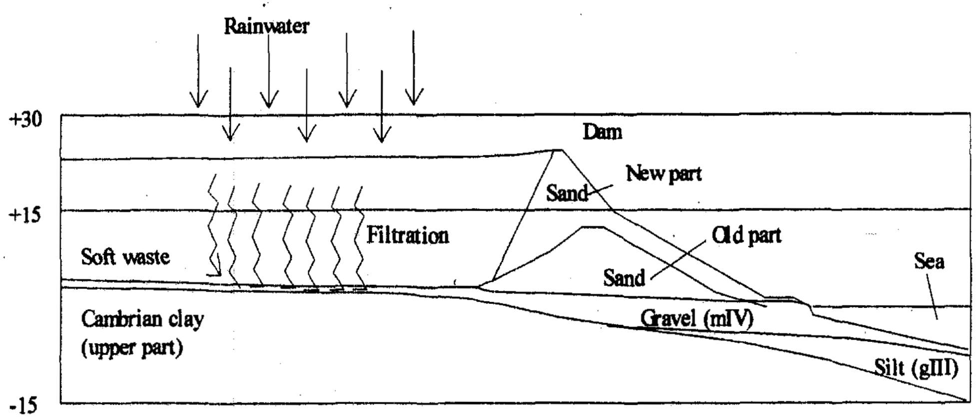

FIGURE 2. Geological section of the tailings pond.

The marine components of the body of the dam are sands, which are loose or medium dense, not having been compressed in the course of the dam's construction. Fine or silty sands are predominant, with some silt. The layer of marine sand or pebbles is a floor of a seam. Gravel and gravel sand are present in the upper part of the dam, which was built later. The total thickness of the layers depends on the dam's height and does not exceed 25.7 m.

Ash occurs as thin layers with varying properties in the body of the dam and in the sediments. The layers were deposited by water transport of slurries, with the water draining out of the layers. The shapes and consistencies of the layers are irregular, having been determined by the placement of the pipes and the nature of the slurries deposited. Some ash layers are cemented, and others are granular material. Maximum thickness of the layers does not exceed 10.7 m. The ash is very sensitive to water and temperature; becomes wet very easily, and when dry becomes crumbly.

Clays and loams fill a part of the pond, forming a weak mass of flowing consistency. More clay is present in the deeper portions of the deposits. The upper layer of 3.6-5.7 m thickness contains coarser fractions. The total thickness of the layer is 10-20 m.

4. Hydrogeological Conditions

4.1 NATURAL WATER

Two hydraulically connected Quarternary and Cambrian-Ordovician water-bearing strata are present in the region. The water in the upper Quarternary stratum may conditionally be divided into soil water and anthropogenic water.

The horizon of the soil water lies in the marine sand and the pebble layer at 0-3.5 m depth from the ground surface and has a free surface. This horizon is fed by precipitation, anthropogenic water percolating through the pond, and the water of the Lower Cambrian-Ordovician stratum. Outflow is into the sea. The chemical composition of the water depends on the composition of the anthropogenic water deposited in the pond, but apparently natural components are also present. The apparently natural ions include carbonate, NH_4^+, Na^+, K^+, HCO_3^-, NO_2^-, and NO_3^-. In addition, the content of Mg^{2+} ions decreases from the northeast to the west, and the content of Ca^{2+} and Cl^- ions is constant across the pond [10]. The pH of this water is >9. Solids content is 10426-21208 mg/l, which is 20-40 times the background for soil water and 3-7 times the background for sea water. Approximately 16-34 tons of solids flow into the sea daily. The blue clays of the Lontova layer are watertight. The horizon is in hydraulic connection with the Lower Cambrian-Ordovician water stratum. The inclination of the water surface toward the sea is 0.04.

In the limestone shore area and the limestone plateau, the Cambrian-Ordovician water stratum lies in the lower Ordovician limestones and Cambrian-Ordovician sandstones, over the watertight blue clays of the Lontova layer.

In 1990, the water table was at 0.4 to 16.9 m depth from the ground surface, at absolute heights of 33 to 38 m. The water stratum has a free surface, is fed by precipitation water, and is hydraulically connected with the soil water table. It contains much less mineralization than the Quaternary water stratum, primarily sulfate, bicarbonate, calcium, and magnesium. The water is moderately aggressive to concrete. In the limestone shore area and the limestone plateau, the inclination of the water surface toward the sea is 0.2-0.3.

4.2 ANTHROPOGENIC WATER

Anthropogenic water is found only in the vicinity of the tailings pond, where it mixes with soil water. The chemical composition of the anthropogenic water varies with the composition of the wastes deposited in the tailings pond.

The hydrogeological situation in the vicinity of the tailings pond has been modified by the presence of the pond and its modifications. Most of the pond is covered with water. This water percolates through the waste into the sea. In the parts of the waste consisting of a mixture of oil shale ash and weak clayey soil, there is no stable water horizon. The conductivity of this layer is small (0.1-0.01 m/day), but other parts of the tailings have much higher conductivity.

For the tailings pond as a whole, the conductivity allows the total precipitation (~600 mm/year) minus evaporation (~300 mm/year), plus the water from the compression of the soft soils, to percolate through to the sea via the marine sand and gravel strata. An approximate calculation gives 152,500 m^3/year of water flowing through the tailings pond and into the sea. Of this amount, 150,000 m^3 is precipitation water, and 2500 m^3 is soil water.

Such a lack of retention of water is favorable for the stability of the dam by minimizing the forces exerted on the dam by the water. Accumulation of water behind the dam would destabilize the dam. However, it is very difficult to block the water flow through the sand and gravel layers.

5. Geotectonic Conditions

The surface of the land is slowly rising (0.2 mm/year) at Sillamäe in response to the removal of the last glaciation. The shore is rising, and erosion will increase. As a result of these processes, sliding forces will develop in the dam and the importance of the counterbalance block will be reduced.

6. Geotechnical Conditions

6.1 SOIL PROPERTIES

Soil properties have been investigated [2, 3, 5, 6, 8, 10, 11, 13, 14, 15]. Static cone penetration tests (CPT), dynamic penetration tests (DPT), plate load tests, and pressiometrics [6] have also been used. Natural soils have also been investigated by dynamical penetration, and soil samples have been taken for laboratory studies [20]. The laboratory studies are not completed. Calculation parameters for the soil are given in Table 1.

TABLE 1. Calculation parameters for soils

Soil	Geol. index	γ_n (kN/m^3)	ϕ (°)	c (kPa)	E_0 (MPa)	K (m/s)
Tailings pond and dam						
Sand	tIV	21.0	32	0	30	0.003
Grav. sand	tIV	21.0	34	0	30	0.003
Ash	tIV	20.0	0	100	25	$3E10^{-5}$
Silty sand	tIV	19.0	0	5	0,5	$3E10^{-7}$
Silt	tIV	18.0	0	15	10	$3E10^{-7}$
Natural soils						
Sand	mIV	20.5	33	0	20	0.005
Gravel	mIV	21.5	38	0	35	0.01
Silty sand	gIII	21.5	0	50	10	-
Weathered clay	Cm_1	20.0	0	80	10	-
Clay	Cm_1	20.5	0	90	15	-

Additional geotechnical investigations (DPT and uniaxial compression strength) showed that the Lower Cambrian clays (Cm_1) underlying the dam differ from classical blue clays on the north Estonian shore in lithological composition as well as mechanical properties. The clay's color is variable (from light blue to dark lilac), it contains considerably more clay, and there are none of the strong thin sandstone layers characteristic of the Cambrian massif. Because the area under investigation lies on the slope of an ancient valley filled with Quaternary sediments, the clay has been subject to glacial and chemical decomposition processes.

The water content of the upper part (15 m) of the blue clays is relatively high, up to 29%. The change in natural water content with depth is shown in Fig. 3, and the dependence of dynamic specific resistance (by DPT) on depth is shown in Fig. 4. The uniaxial compressive strength of the soil is 70-230 kPa. The uniaxial compressive strength of the clays in the upper part is 20-25% of this. The water content of the clay decreases somewhat with depth (Wn = 15-20%), and the compressive strength increases by 100-250 kPa. In the area under investigation, the strong Cambrian clay begins at a depth of 20-25 m.

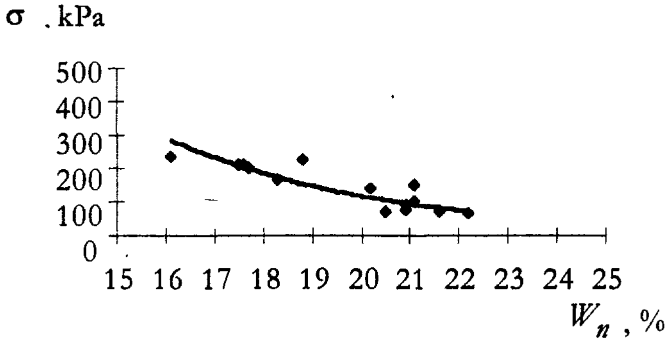

FIGURE 3. Dependence of uniaxial pressure strength of Cambrian clays on natural water content.

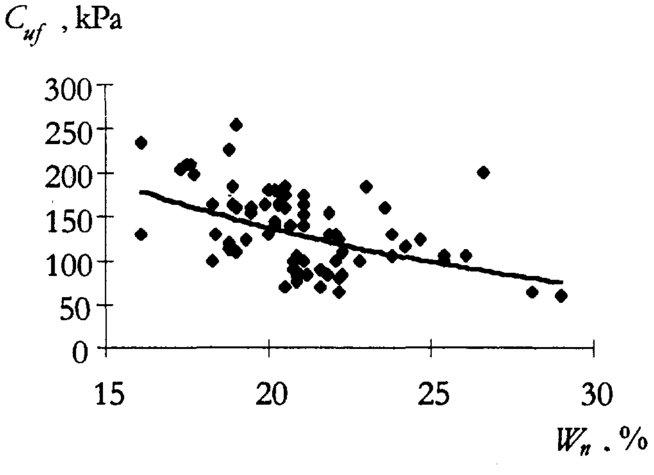

FIGURE. 4. Dependence of shear strength of Cambrian clays on natural water content.

Investigations of slope stability at the Tallinn Kopli clay pit gave similar results. Clays with greater moisture content than usual and weaker mechanical properties were observed there.

6.2 GEODYNAMIC SITUATION

The stability of the tailings dam is primarily influenced by three closely related processes: slope processes, shore processes, and hydrodynamic processes. Anthropogenic and geotectonic processes are given less weight in this analysis, but they may be taken into account in future analyses.

6.2.1 Slope Processes

The stability of the dam has been discussed in references 13-21. Stability was evaluated (in the case of weak clay and ash layers and blue clays) on the basis of minimum strength parameters in accordance with international practice. An average of plane shear strength tests, the results of CPT tests, was used for sand layers.

The stability of the dam was calculated for 13 profiles. The calculations were made with the Nõlv program, developed by AS GIB, based on the theory of circular cylindrical slickensides. The configuration of the slickensides was varied to give the minimum (worst case) result for each section. The safety factor was determined for more than 400 different forms of slickensides. Hydrodynamic forces were not considered. Slope calculations with a fixed stability for sand (if the slickenside permeates the body of the dam) give a stability factor F = 1.6-3.6, which guarantees the stability of the slopes. Because of the raising of the dam's height, deformations in the body of the dam have already taken place. Thus it may be assumed that initially uncompacted material in the dam became denser as a result of those deformations, providing a new equilibrium.

The slide probability along the weak, saturated, and decomposed surface of the Cambrian clays is small because of the small inclination of the clay's surface. The calculated stability factors are F = 1.3-1.6.

Stability calculations for the deeper layers of the blue clay give safety factors of F = 1.05-1.3. These factors are less than is required by the standards (F > 1.5) to guarantee a stable slope.

The slope calculations were verified by specialists at the Norwegian Institute of Geotechnics.

6.2.2 Shore Processes

The shores of Sillamäe, from Voka settlement in the west to the small cape with a mounded gravel shore on the eastern edge of the town of Sillamäe is subject to active sea erosion. From the west, a section of bank shore is being actively eroded, the terrace shore in front of the tailings pond is being actively eroded, the shore along the center of the town is partly protected against erosion, and a bathing-beach is subject to intense erosion up to the mounded gravel shore.

Erosion has become more severe since 1993. Erosion is especially intense at the northern part of the tailings dam. After every storm, significant removal of material at the foot of the dam is observed. The northeastern shore of the dam is better protected against erosion by rocks at the foot of the erosion terrace. The shore in the eastern part of the town of Sillamäe has been severely eroded by storms.

Investigations of shore processes in the Sillamäe coastal area, among them observations made in spring and autumn of 1994, showed that the whole coast has not gained dynamic stability. A large number of cyclonic storms have resulted in the expansion of eroded areas and the intensification of erosion processes which can be expected to continue for the next several decades. Human activities could also contribute to further erosion of the gravel terrace supporting the tailings pond.

6.2.3 Hydrodynamic Processes

The tailings pond now presents a fairly stable hydrodynamic situation. In effect, the pond acts as a filter for materials deposited in it. The percolation coefficient of the solids in the pond is small, between k = 0.01 and k = 0.1 m/day. The percolation coefficient of the dam is 10 m/day, and of the natural gravel layer under the dam, 40 m/day. This hydrodynamic scheme practically guarantees the vertical movement of water through the wastes and the body of the dam and into the sea through the gravel layer below the pond. Hydrodynamic forces on the body of the dam are practically zero. This regime has been present for a long time. Reduction of the outflow of water would increase the horizontal forces on the dam and destabilize it.

7. Geotechnical Surveillance

7.1 OBSERVATION OF SHORE PROCESSES

For calculating the stability of the dam and blue clay massif, the sea, sea-bottom sediments, and coastal sediments are the counterbalance. They remain in the outpressure area of the slickenside and improve the stress situation, providing a higher safety factor with a larger counterbalance. Thus the severe shore erosion is a particularly significant factor, because it reduces the counterbalance. This process could cause dam failure long before waves actually touch the dam.

Surveys of the shore in 1994 and 1995 showed that erosion had become more rapid and the mass of gravel in the counterbalance body had been significantly reduced. No investigations of shore processes are currently in progress for the Sillamäe area. A continuing program of observations is needed.

7.2 MEASUREMENTS OF DAM DEFORMATION

A network of benchmarks was established in the summer of 1994 for measuring the deformation of the dam. Five measurement cycles have been made since then, in July and November 1994, May and December 1995, and August 1996. The network consists of 22 benchmarks, of which two are deep benchmarks (length 24 m), nine are high soil benchmarks with lengths of 6 m, and 11 are low soil benchmarks with lengths of 3 m. The foundation depths are 1-2 m less than the lengths. Figure 5 is a map of the locations of the benchmarks.

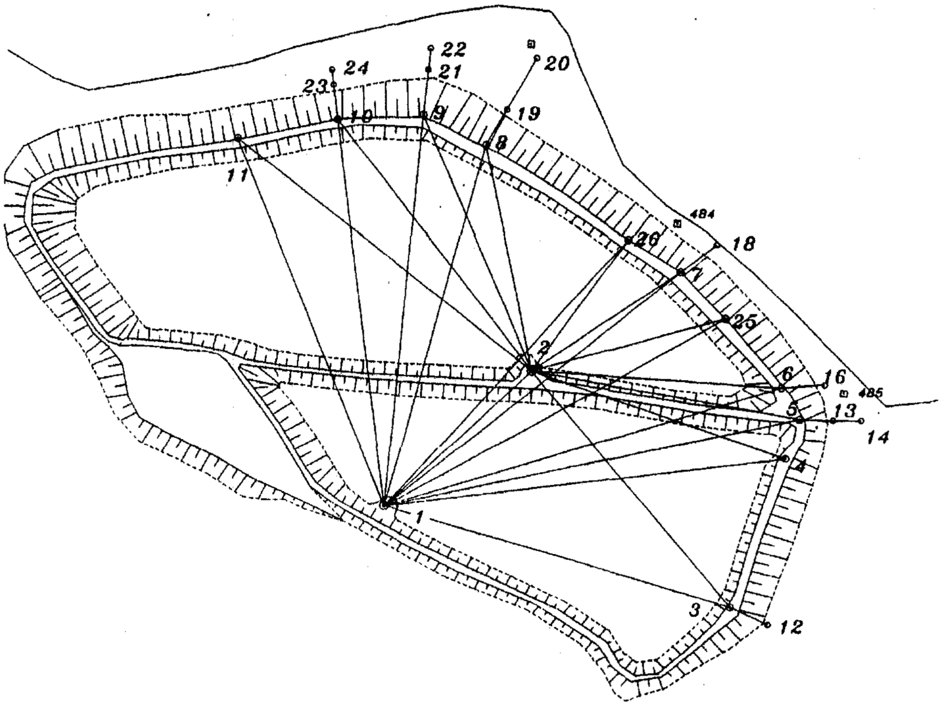

FIGURE 5. Map of benchmark locations.

Two measuring systems, laser and global positioning system (GPS), are used to confirm measurements. The horizontal stability of the deep benchmarks is assumed as a result of their placement outside the slide area. All bench marks placed on the upper edge of the dam are measured by the laser instrument on the basis of the two deep benchmarks. Distances to and from the benchmarks are measured three times and the median is calculated. GPS measurements are based on the Navstar system of the US Department of Defense.

The measurements are presented in Figs. 6 and 7. The results confirm the existence of marked deformations on the seaside edge of the dam. The positions of benchmarks 3-6, in the northeastern part of the dam, move vertically 2-5 mm/year, and horizontally 28-32 mm since the measurements began. Benchmarks 8-11, in the northwestern part of the dam, move vertically 10-30 mm/year. Benchmarks 8 and 10 have moved horizontally a total of 41 mm toward benchmarks 1 and 2 since the measurements began, but the deformation speed has slowed from 38 to 22 mm/year. Benchmarks 9 and 11 initially moved away from benchmarks 1 and 2 by 15 mm, but the direction of movement has changed, and the rate of movement is now 8 mm/year. If the movement of benchmarks 8-11, on the northern side of the dam, is south, then benchmarks 3-7, on the eastern side of the dam, have moved east. The deformation speed for these benchmarks ranges from 3 to 30 mm/year. No benchmark is moving faster than 33 mm/year.

Benchmarks 19, 20, 23, and 24, in front of the tailings pond (beach side), have shown no vertical movement. However, their horizontal distances from benchmarks 1 and 2 have lengthened by 25-54 mm. There has been no horizontal movement of benchmarks 21 and 22 (Fig. 7). Benchmarks 12 and 14, at the base of the eastern part of the tailings pond have moved 7-9 mm to the east, with no vertical movement.

The observed movements of the benchmarks indicate two kinds of slope processes. One is a deep creep involving the blue clay massif under the northern and eastern parts of the tailings pond, and the other is local creeping inside the body of the dam. On the basis of the existing information, the use of inclinometers for measuring movement would require surveillance to determine the optimum placement of the points at which measurements are to be made.

7.3 SURVEILLANCE OF PORE PRESSURE

Measurement of pore pressure allows observation of stresses in the soil in otder to evaluate slope stability and deformations in deeper layers, contributes information for the calculation of slope stability, and provides a warning of dangerous deformation.

In cooperation with the Norwegian Geotechnical Institute, piezometers were placed in and around the tailings pond in April 1996, to measure the changes in pore water pressure of the soils and changes in stresses with time. Five pick-offs were placed. Measurements are made once a month. Results are shown in Fig. 8. The benchmarks as well as the pore pressures indicate an unstable situation in the strata under the dam. In deeper strata (Cambrian clay), the pressure falls as much as 5 kPa in a month, indicating the deformation of a slickenside. In the upper soil layers (Upper Cambrian clay, moraine), no definitive changes are observed.

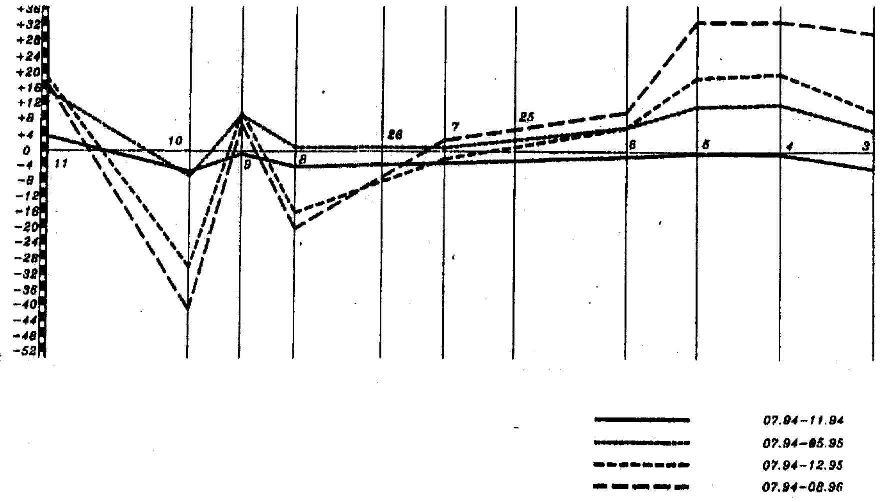

FIGURE 6. Results of the deformation measurements on the dam.
Horizontal deformations at the upper edge of the dam.

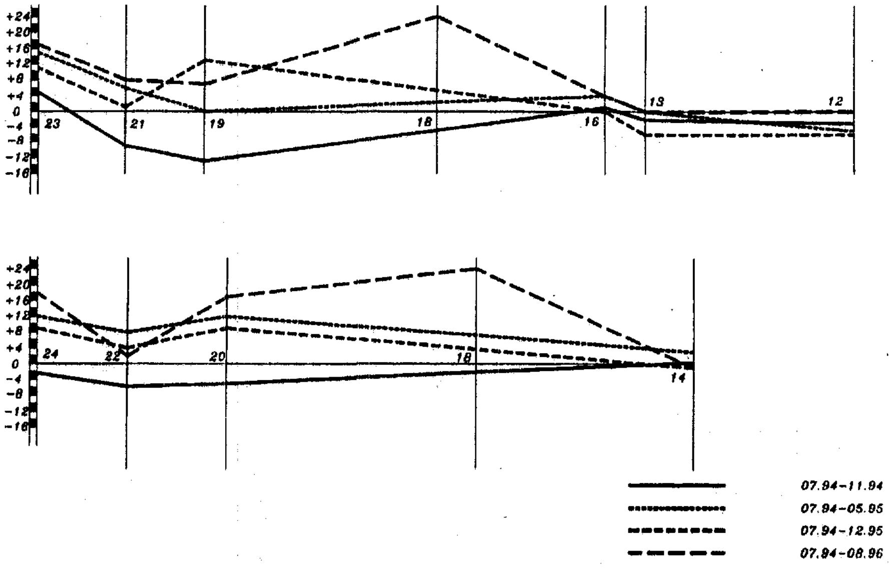

FIGURE 7. Results of deformation measurements on the dam. Horizontal deformations at the base of the dam.

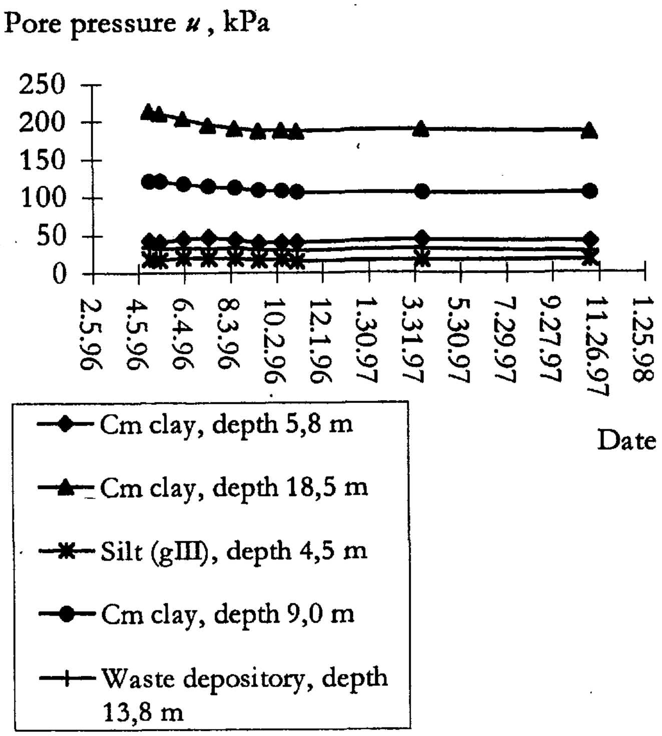

FIGURE 8. Results of monitoring of pore pressure

7.4 ENVIRONMENTAL SURVEILLANCE

Continuing measurements of the release of contaminants are described in V. Nosov's paper in this collection [25]. Radiation levels are low enough that they should have negligible effect on the stability of the dam. An issue that needs to be investigated is the chemical effect of the contaminants on the condition of the soil, particularly on the strength of the blue clay. Changes in the chemical conditions of soil and consequently in its strength parameters can take place within years.

8. Summary

In its present condition, the dam of the tailings pond is less stable than is recommended by international practice. A surveillance system has been established to observe both vertical and horizontal soil movement and the buildup of stresses in the soil. The stability of the dam depends primarily on erosion by shore processes and the consequent reduction of the counterbalance, as well as the reduction in strength of the Cambrian clay underlying the dam caused by creep and by water percolating to the bottom of the pond. The most critical need is to enlarge the counterbalance and to stop erosion. The surveillance system should be supplemented with a system of inclinometers. Monitoring of coastal processes should be continued.

At present, the hydrodynamic stress on the dam is minimal because the pebble layer allows drainage of water out of the tailings pond and into the sea. The regulatory requirement to end emissions into the Baltic Sea must be met without causing undesirable hydrodynamic stresses on the dam. This could be done by covering the pond and diverting precipitation water.

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PROCESS FLOW SHEET

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1. Introduction

At present, production at AS Silmet consists of three parts: rare earths, rare metals, and metallurgical. The technology used in production is primarily hydrometallurgical, including dissolution, precipitation, filtration, and liquid extraction processing. The final stages of production include drying and calcination in electric drum furnaces. All production wastes are neutralized and sent to the tailings pond.

2. Rare Earth Production

2.1 RAW MATERIALS

The raw material for this production is the chloride melt of rare earth elements from the Solikamsk Magnesium Plant. The chloride melt contains 38-42% rare earths, mainly of the light group (calculated as Ln_2O_3 oxides). The melt is obtained by chlorinating loparite concentrate. The composition of the rare earth metals in the melt corresponds to their natural composition in loparite (Table 1).

TABLE 1. Rare Earth Composition of Chloride Melt (percent of the sum of the oxides)

Ln_2O_3	26.8	Tb_4O_7	0.017
CeO_2	53.8	Dy_2O_3	0.051
Pr_6O_{11}	5.1	Ho_2O_3	0.0043
Nd_2O_3	13.2	Er_2O_3	0.0035
Sm_2O_3	0.88	Tm_2O_3	0.0003
Eu_2O_3	0.134	Yb_2O_3	0.00036
Gd_2O_3	0.185	Lu_2O_3	0.00012
Y_2O_3	0.025		

Rare earth metals in the raw material are contaminated by admixtures (calcium, strontium, sodium, potassium, iron, aluminum, silicon), as well as radioactive elements (uranium, thorium) and their decay products. The overall chemical composition of the chloride melt is shown in Table 2.

TABLE 2. Overall Chemical Composition of Chloride Melt (%)

Ln_2O_3	38-42	Na_2O	7-8
U_3O_8	0.01	K_2O	0.2-0.3
ThO_2	0.15-0.72	Fe_2O_3	0.1-0.2
CaO	5-6.2	SiO_2	2.5-3.2
SrO	2.1-2.7	Cl	44-52
BaO	0.13-0.15		

2.2 OUTLINE OF PROCESS

The outline of the production process is shown in Figure 1 and consists of the following main stages:

2.2.1 Dissolution

The chloride melt is dissolved in river water with active stirring and air is blown through to promote oxidation of iron.

2.2.2 Deactivation 1

Ammonium sulfate solution is added to the slurry to precipitate strontium and calcium sulfates. Most of the radium is also precipitated as the sulfate.

2.2.3 Thorium Hydroxide Precipitation

Thorium is precipitated by use of an ammonia solution at a pH of 3.8 to 4.1with heating. In addition to the thorium hydroxide, iron hydroxide is precipitated along with aluminum, uranium, and a small amount of the rare earth elements.

2.2.4 Filtration

After precipitation, the slurry is filtered. Solids consisting of undissolved melt, calcium sulfate, strontium sulfate, and the hydroxides are washed with water to remove solution and are discharged into the tailings pond as a water slurry. The washing water is used to dissolve the melt and to dilute the slurry. The main filtrate goes to Deactivation 2.

2.2.5 Deactivation 2

A solution of barium chloride is added to the rare earth solution, which contains excess sulfate. The remaining radium in the solution is precipitated with barium sulfate. The slurry is again filtered. The solids are combined with the thorium hydroxide residues. The deactivated solution is sent to the next step in the processing.

2.2.6 Precipitation of Carbonates

The rare earth carbonates are precipitated by use of a solution of NH_4HCO_3 and $(NH_4)_2CO_3$. This separates them from the admixture elements. The solid is filtered, and the carbonates are washed with water. The filtrate and wash water are combined and discharged to the tailings pond. The washed carbonates may be used as a final product or as a material for further processing.

The carbonates are calcined in rotating electric furnaces to produce oxides. For fluorides and polishing powders, the carbonate water slurry is fluorinated using a

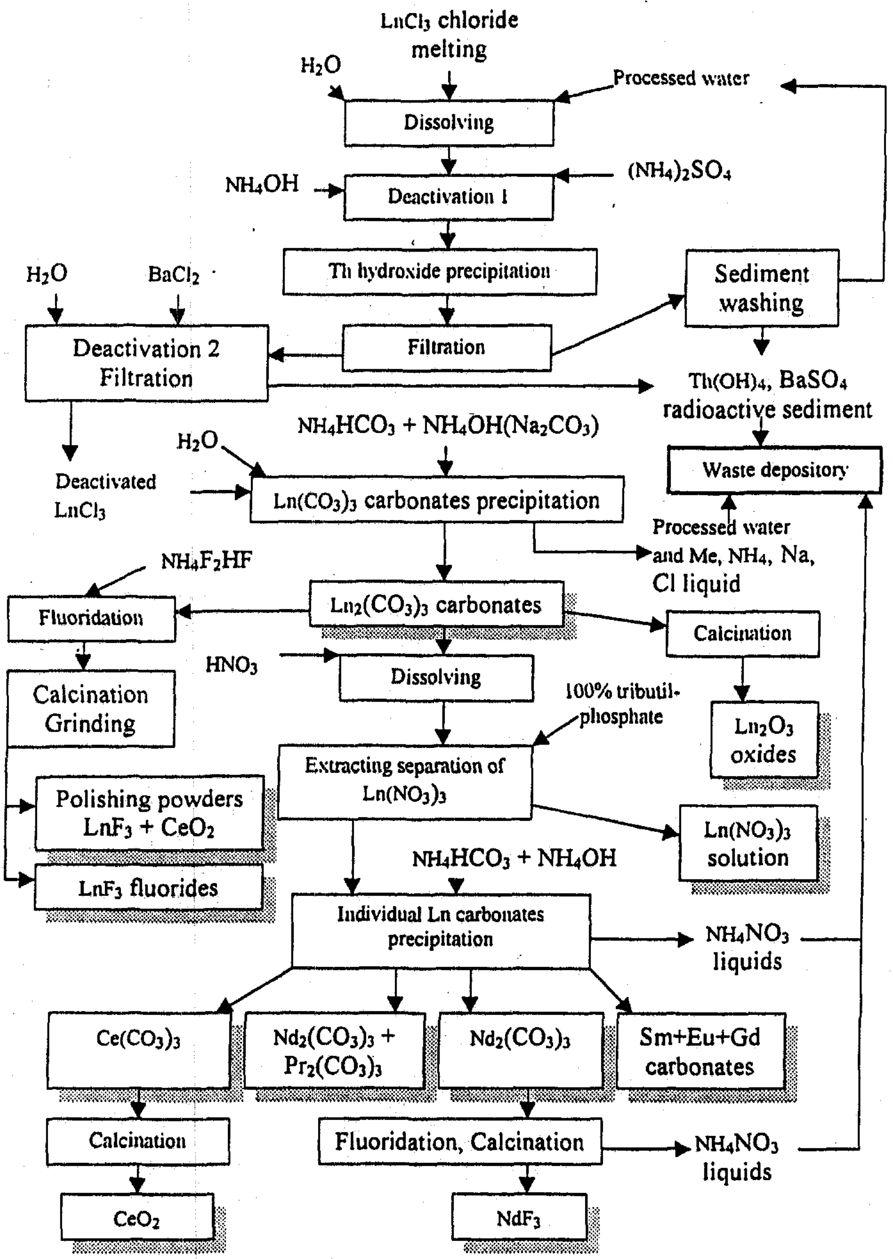

FIGURE 1. Flow Sheet of Chloride Melt Processing for Rare Earth Production.

solution of ammonium fluoride-bifluoride. In order to obtain individual rare earth elements, the carbonates are dissolved in nitric acid and subjected to liquid-liquid extraction.

2.2.7 *Separation by Extraction*

Separation of the rare earth elements is performed from solutions weak in acid (15-30 g/liter of HNO_3) but concentrated in the rare earths (350-380 g/liter Ln_2O_3). The extractant is 100% tributyl phosphate. Extraction takes place in a continuous counterflow in mixer-settler extractors with sectioned stratification chambers. Water is used for re-extraction to recover the individual rare earths. Carbonates of individual elements are precipitated from the re-extracts and are used as the final products or as materials for producing oxides or fluorides. Spent nitric acid solutions are discharged to the tailings pond.

2.3 WASTES FROM THE PROCESS

The major chemical reagents used in the process are H_2SO_4, HNO_3, NH_4HCO_3, NH_4F*HF, Na_2CO_3, $BaCl_2$, and tributyl phosphate. As a result, different kinds of waste are produced. The composition of the different types of waste and the amounts for one ton of raw material processed are given in Table 3.

2.3.1 *Radioactive Waste*

This is obtained as a liquid slurry after the deactivation of the chloride solutions. The solid phase of the slurry is a mechanical mixture of insoluble substances contained in the chloride melt (loparite, erigine), calcium sulfate; strontium, barium, and coprecipitated radium sulfates; and thorium, iron, aluminum, and rare earth hydroxides.

The liquid phase of the slurry is dilute ammonium chloride solution contaminated by soluble chlorides of alkaline, alkaline earth, and rare earth elements. In our opinion, the best way to reduce and decontaminate this kind of waste is to change the deactivation technology. For example, soda might be used in the Deactivation 1 and Thorium Hydroxide Precipitation steps (Figure 1) in place of ammonium reagents, which would prevent ammonium ions from appearing in the liquid phase of the slurry and the rare earth solutions. Another possibility is to remove the components that appear in the solid phase of the slurry from the process separately and to develop decontamination and storage technologies for each of them.

2.3.2 *Chloride Liquids and Process Water*

This dilute solution of ammonium chloride containing water-soluble chlorides of alkaline, alkaline earth, and rare earth chlorides results from the precipitation and washing of the combined rare earth element carbonates. Again, soda could be substituted for the nitrogen-containing precipitant. This would result in the discharge of sodium chloride solutions into the Baltic Sea from the tailings pond.

TABLE 3. Composition and Amounts of Different Types of Waste for One Ton of Raw Material

Waste	**Amount from one ton of raw material**	**Components Content**			
Radioactive waste as a slurry	0.37 m^3		Solid, %		
Solid	0.06 ton	Ln_2O_3	11.6-12.3	BaO	9.8-10.2
		U_3O_8	0.14-0.16	Fe_2O_3	1.9-2.0
		ThO_2	9.6-11.1	SiO_2	6.0-7.0
		CaO	0.2-0.3	SO_4	12.4-14
		SrO	6.8-7.0		
Liquid	0.32 m^3		Liquid, g/l		
		Ln_2O_3	2.0-2.5	K_2O	0.015
		CaO	0.3	Cl	2.6
		SrO	0.1	NH_4	0.3-1.0
		Na_2O	0.3		
Chloride	20 m^3	Ln_2O_3	0.2-0.3	Na_2O	10-13
		CaO	2-3	Cl	12-14
		SrO	1-1.3	NH_4	0.7-0.8
Nitric liquids and process water	10 m^3	NH_4	10-15	NO_3	40-50
Fluoride liquids	0.4 m^3	F	7-10	HCO_3	10-12
		NH_4	13-16	CO_3	5-7
Solution from regeneration *of extractant*	0.2 m^3	Na_2O	26-30	NO_3	70-80
		Tributyl phosphate	0.1-0.3	$Na_2C_4H_9PO_4$	0.2-0.5

2.3.3 *Nitric Liquids and Process Water*

This dilute ammonium nitrate solution is the major liquid waste from the Silmet plant contaminating the Baltic Sea with nitrogen. It is obtained during precipitation of the separated carbonates from the nitric re-extracts.

Again, soda could be substituted for the ammonium carbonate precipitating reagent to remove the ammonium component of this solution. Other ways to discontinue release of ammonium nitrate to the Baltic Sea are production of ammonium nitrate for fertilizer or explosives, recycling of the nitric acid, or decomposition of the nitrate by a plasma or burning in a furnace.

2.3.4 *Fluoride Liquids*

This solution contains fluoride and ammonium ions and is obtained during fluoridation of the carbonates by ammonium fluoride-bifluoride. This waste is produced in small amounts and could be neutralized with ash and disposed of in the tailings pond, where the ammonium would be removed naturally.

2.3.5 *Solution from Regeneration of Extractant*

This solution contains sodium nitrate, butylphosphoric acids, and tributyl phosphate and is obtained from the purification of the tributyl phosphate. This waste might be decontaminated by decomposition of the organic components to phosphates with subsequent denitrification.

3. Rare Metals Production

3.1 RAW MATERIALS

The production of the rare metals niobium and tantalum uses columbite concentrates, whose composition varies depending on their place of origin. Contaminants in the concentrates include iron, tin, titanium, manganese, radioactive elements (uranium, thorium), and their decay products. The chemical composition of the concentrates is shown in Table 4.

TABLE 4. Chemical Composition of Concentrates (%)

Nb_2O_5	58-60	MnO_2	1.7-4.0
Ta_2O_5	5.8-6.1	WO_3	1.1-0.5
TiO_2	5-7	P	0.4-0.1
Fe_2O_3	20-18	CaO	0.05-0.1
Al_2O_3	0.4-1.0	ThO_2	0.38-3.0
SiO_2	1-2	U_3O_8	0.085-0.2
SnO_2	2.6-5.9		

3.2 OUTLINE OF PROCESS

The outline of the process is presented in Figure 2 and includes the following major stages.

3.2.1 Grinding

The concentrate is ground in circular mills (ball mills?) to a size of 0.3-0.16 mm.

3.2.2 Dissolution

The pulverized concentrate is dissolved in a mixture of concentrated acids (92.5% H_2SO_4 and 40% HF). The dissolution is performed batchwise in reactors with mechanical stirring. The concentrate is dissolved in small portions, because the process is highly exothermic, heating the slurry to 70-85 C. Emerging gases are condensed in cooled heat exchangers, and fluoride is removed by soda.

3.2.3 Filtration and Washing of Solids

After dissolution, the slurry is filtered. The insoluble remnant is washed with dilute sulfuric acid. The wash waters are combined with the main filtrate; the free fluorine content is corrected, and the liquid is sent to the extraction step. The washed solids are sent to the tailings pond as a slurry.

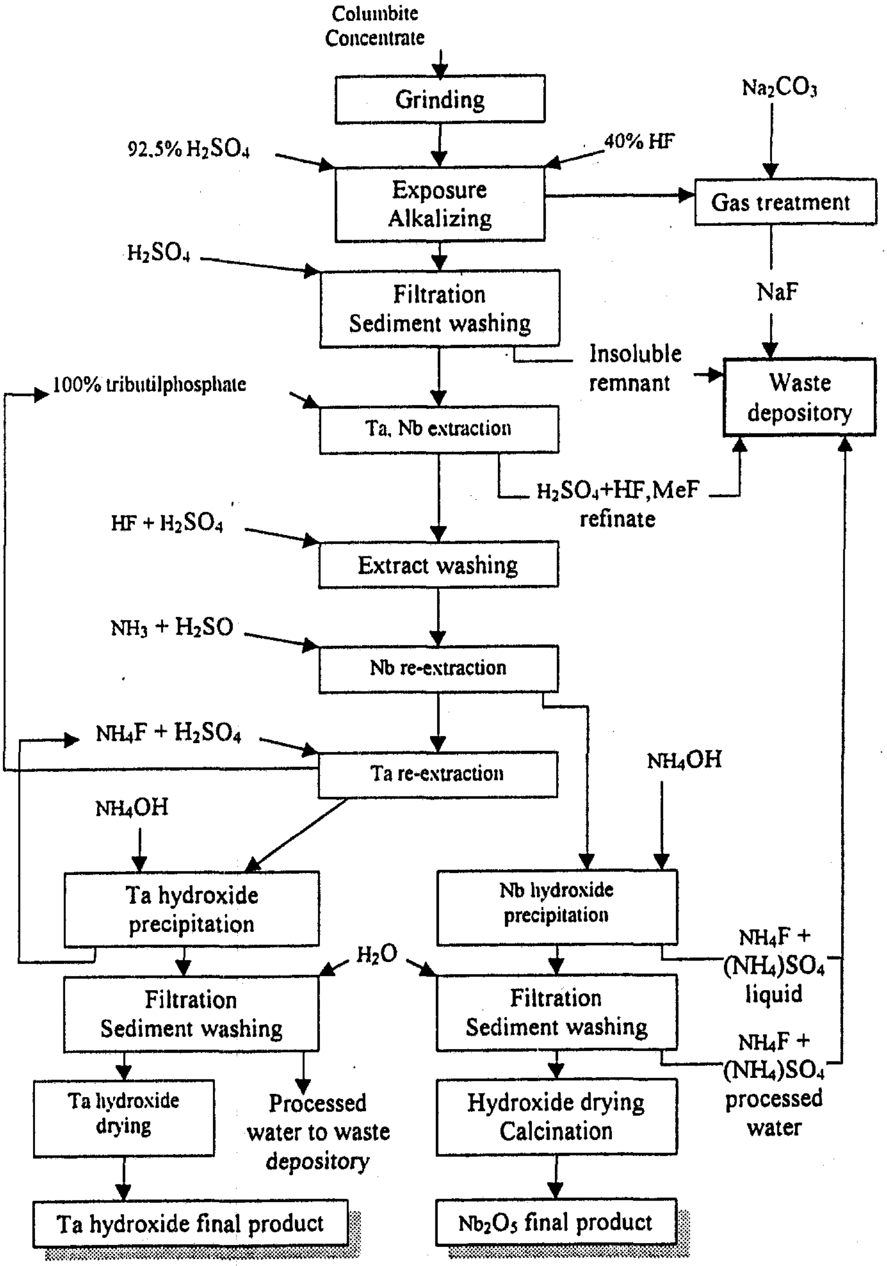

FIGURE 2. Flow Sheet for Processing of Columbite Concentrates for Rare Metals Production.

3.2.4 Extraction of Ta and Nb

Tantalum and niobium are extracted from the H_2SO_4 + HF solution by 100% tributyl phosphate in extractors of the mixer-settler type. Tantalum and niobium are extracted together into the organic phase, and the aqueous raffinate is discharged to the tailings pond.

3.2.5 Extract Washing

The tantalum and niobium extract is washed with a solution of H_2SO_4 + HF. The wash waters are combined with the aqueous raffinate from the extraction, and the washed extract is sent to the re-extraction step.

3.2.6 Re-extraction of Nb

Niobium is selectively re-extracted with an ammonium sulfate solution. From the re-extracts, tantalum is extracted with a small amount of tributyl phosphate. Then niobium hydroxide is precipitated with ammonia. After precipitation and filtration, liquids and wash waters are discharged to the tailings pond. The washed hydroxide is dried and calcined to produce the 5-oxide as a final product and raw material for metallurgical production.

3.2.7 Re-extraction of Ta

Tantalum is re-extracted with a recycled solution of ammonium bifluoride. Tantalum hydroxide is precipitated from the re-extracts by ammonia; the precipitate is filtered and washed of solution. After precipitation, the liquid solution is recycled. Process water is discharged to the tailings pond. The washed hydroxide is dried for the final product.

3.3 PROCESS WASTE

In rare metal production, the major reagents are sulfuric and hydrofluoric acids, ammonia water, soda, and tributyl phosphate. Several kinds of process waste are obtained. The amounts and composition of these wastes for one ton of raw materials are shown in Table 5.

3.3.1 Insoluble Remnant

This is obtained as a slurry after dissolving the columbite concentrates and after filtering and washing. This solid phase is a mixture of insoluble chemical compounds of various elements (oxides, fluorides, sulfates) of various elements, mainly iron, tin, manganese, and thorium.

The liquid phase is a dilute solution of sulfuric and hydrofluoric acids contaminated by water-soluble salts (sulfates and fluorides) of various elements.

A possible method of preparing this insoluble remnant for disposal is washing it to remove water-soluble salts, followed by cementation. It is possible to precipitate insoluble compounds of metals, calcium sulfate, and fluorides from the washing solution and slurry by using ashes. The resulting precipitates can be cemented and disposed of as solid waste, and the resulting liquids could be discharged into the sea.

TABLE 5. Composition and Amounts of Different Types of Waste for One Ton of Raw Material

Waste	Amount from one ton of raw material	Components Content			
Insoluble remnant as a slurry	1.1-1.2 m^3	Solid, %			
Solid	0.297 ton	Nb_2O_5	4.9	ThO_2	1.3
		Ta_2O_5	0.7	SiO_2	1.0
		Fe_2O_3	44-45	P	0.7
		SO_4	27-28	TiO_2	4.1
		SnO_2	8.4	U_3O_8	0.3
		MnO_2	3.7	CaO	0.16
		WO_3	2.4	F	10-19
Liquid	0.32 m^3	Liquid, g/l			
		F	25	H_2SO_4	80-100
Raffinate after extraction	5-6 m^3	Nb_2O_5	0.1	Al2O3	0.8
		Ta_2O_5	0.01	SiO_2	1.8
		Fe_2O_3	17-18	P	0.4
		TiO_2	7.6	H_2SO_4	380
		SnO_2	0.2	NH_4	9.4
		MnO_2	1.2	F	74-76
		WO_3	0.8		
Chloride	20 m^3	Ln_2O_3	0.2-0.3	Na_2O	10-13
		CaO	2-3	Cl	12-14
		SrO	1-1.3	NH_4	0.7-0.8
Nitric liquids and process water	10 m^3	NH_4	10-15	NO_3	40-50
Fluoride liquids	0.4 m^3	F	7-10	HCO_3	10-12
		NH_4	13-16	CO_3	5-7
Solution from regeneration of extractant	0.2 m^3	Na_2O	26-30	NO_3	70-80
		Tributyl phosphate	0.1-0.3	$Na_2C_4H_9PO_4$	0.2-0.5

3.3.2 Raffinate

This solution of sulfuric and hydrofluoric acids contaminated by water-soluble salts is obtained during the extraction of tantalum and niobium. Possible methods of decontamination include removal of fluoride by chemical means or by burning, production of sulfate fertilizers, precipitating salts into an insoluble form and disposal as solid waste, and treatment of liquids to acceptable compositions for disposal in the sea.

3.3.3 Liquid Solutions and Process Water after Niobium Hydroxide Precipitation

This dilute solution of ammonium sulfate and fluoride is obtained during precipitation and washing on the hydroxide. The main problem in decontaminating this kind of waste is the nitrogen present as ammonium ion.

A possible method of handling is to separate the dilute process water stream from the fluoride-containing liquids (concentrations of 200 g/liter). The more concentrated fluoride-containing liquids can be treated for fluoride removal, and the process water can be treated to remove fluoride, sulfate, and ammonia.

3.3.4 Process Water from Tantalum Hydroxide Precipitation

This dilute solution of ammonium fluoride is obtained during washing of the hydroxide. It can be treated with the washing waters from niobium hydroxide.

3.3.5 Solutions from Extractant Regeneration

These solutions are obtained from purification of the tributyl phosphate extractant with soda solution to remove butylphosphoric acids. These solutions contain sodium salts of butylphosphoric acids, excess soda, and dissolved tributyl phosphate. These solutions might be treated to decompose the organic components to phosphates, which would then be precipitated.

4. Metallurgical Production

The raw material is the niobium pentoxide from the rare metals production. The process steps are reduction to metal in furnaces using an alumocalciothermal method, and remelting of niobium in electron-beam furnaces for purification.

The waste is the dross after the reduction stage. It is water-insoluble and is currently being disposed as a solid waste. The amount of dross is approximately 0.4 ton for 1 ton of metal product. The chemical composition is 28-33% CaO, 65-75% Al_2O_3, and 1-2% Nb_2O_5. It can also be used to produce aluminum and as a construction material.

RARE EARTH PRODUCTION AT THE SILLAMÄE PLANT, 1969-1991 AND THE POSSIBILITY OF SCANDIUM EXTRACTION FROM LOPARITE

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Loparite concentrate (Table 1) was first processed at Sillamäe in 1969 to recover niobium and tantalum for use in nuclear power. The first batch of rare earths was produced in 1970.

TABLE 1. Composition of Loparite Concentrate [1, 2]

Component	TiO_2	Nb_2O_3	Ta_2O_3	ThO_2	Na_2O	K_2O	CaO	SrO	SiO_2	REO	Sc_2O_3
Content, %	39.24	7.1	0.54	0.57	9.06	0.75	5.26	0.62	0.27	32.29	0.0002

In the 1970s, the supply of loparite concentrate from the Kola Peninsula was irregular (Table 2), and the consumption of rare earths in industry was low, so technical-grade carbonates were stored.

TABLE 2. Delivery of Loparite Concentrate to Sillamäe Plant

Year	1971	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985
Amount, t	7.2	5.6	5.5	2.4	3.6	4.0	4.0	4.5	4.5	5.0	5.1	5.77	5.42	9.6	10.5

In the following years, these stockpiles were worked off along with current processing.

Loparite concentrates were processed by the method of solid-phase sulfation, which was replaced in 1984 by the improved liquid phase sulfation method. As a result, titanium, tantalum, and niobium formed sulfates, but the rare earths were recovered as double sulfates (Figure 1), which have low solubility in acid and nitrate solutions, allowing the separation of these groups of elements [3]. Processing of loparite concentrate by a mixture of sulfuric acid and ammonium sulfate involves the following main processes.

$$\begin{aligned}
&Ta_2O_5 + 3\,H_2SO_4 + (NH_4)_2SO_4 = 2\,NH_4TaO(SO_4)_2 + 2\,H_2O \\
&Nb_2O_5 + 3H_2SO_4 + (NH_4)_2SO_4 = 2\,NH_4NbO(SO_4)_2 + 3\,H_2O \qquad (1) \\
&TiO_2 + H_2SO_4 + (NH_4)_2SO_4 = (NH_4)_2TiO(SO_4)_2 + H_2O \\
&Ln_2O_3 + 3\,H_2SO_4 + (NH_4,K,Na)_2SO_4 = (NH_4,K,Na)Ln(SO_4)_2 + 3\,H_2O
\end{aligned}$$

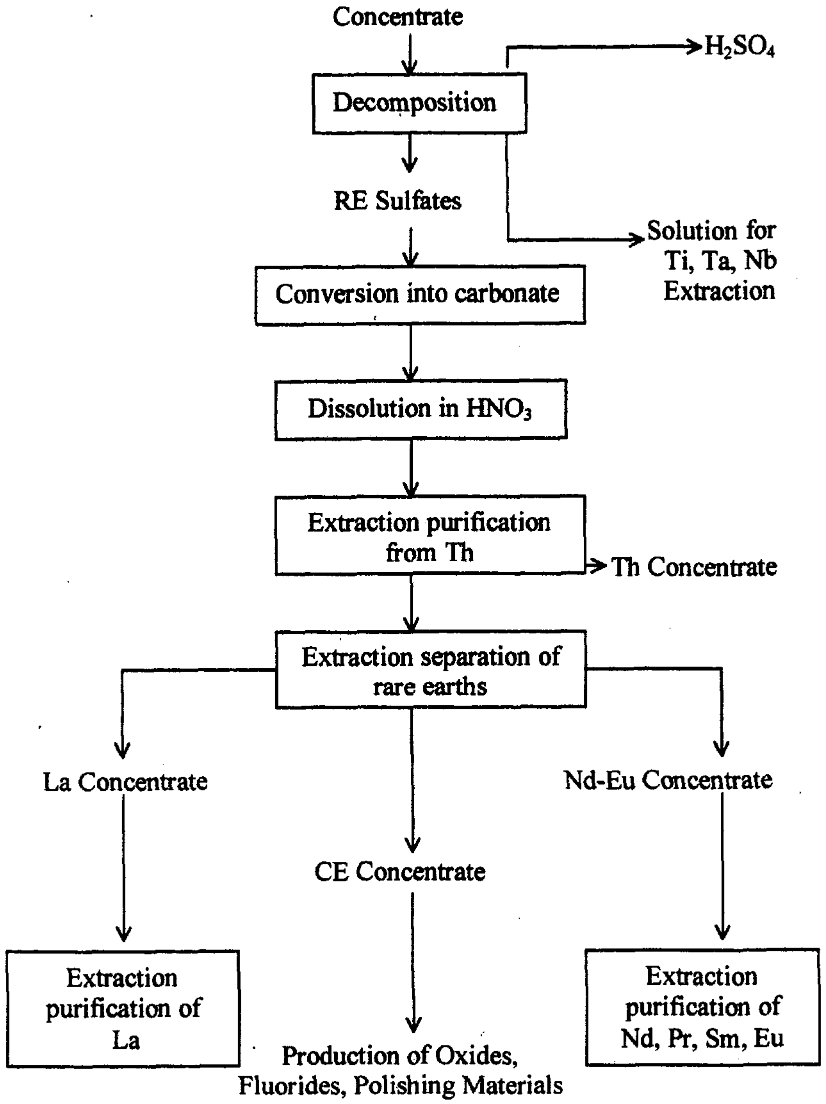

FIGURE 1. Recovery of Rare Earths from Loparite

Double sulfates were the source material for rare earth production. The carbonates were converted within 5 hours at a temperature of 70 C by an ammonium carbonate and hydroxide solution. After filtering, the carbonates were dissolved in nitric acid. The resulting ammonium sulfate byproduct solution was evaporated to dry crystals, which were used as fertilizer on Estonian fields.

The next step is removal of the radionuclides thorium and radium. Thorium is precipitated by ammonium hydroxide at pH 4.8, and radium coprecipitates with barium sulfate. The thorium cake is sent to the tailings pond. Niobium and tantalum are then extracted. At the beginning of the 1990s, thorium removal by extraction with tributyl phosphate was developed [4], but it was not used in the plant.

Removal of the radionuclides by this method is much simpler than the corresponding deactivation for uranium-containing concentrates [5]. It is a safe method for preparing rare earth products that meet health and safety requirements.

Before 1987, the Sillamäe plant produced only mixed rare earth products (Table 3). Then extraction cascades (more than 300 units) were placed in operation. The rare earths were in nitric acid solutions, with tributyl phosphate as the extractant. Shelf-type mixer-settlers with capacities of 1 and 5 m^3/h were used for extraction separation of the individual rare earths. The technology gives concentrates of lanthanum, cerium, neodymium, and the middle group of rare earths up to 99.5% purity.

TABLE 3. Sillamäe Plant Rare Earth Production (tons)

Product Year	Total	Solutions for Oil Catalysis	Oxides for Metallurgy and Export	Fluorides	Polishing Powder	
					Technical	Optical
1972	588	588				
1973	616	616				
1974	637	589	48			
1975	678	500	178			
1976	601	421	190			
1977	837	256	581			
1978	1193	589	473	131		
1979	1325	750	575			
1980	1470	765	605			
1981	1630	620	550	360	100	
1982	1580	330	780	190	150	130
1983	1236*	180	563	82	91	320
1984	1545	219	878	48	115	285
1985	2090	180	1460	50	115	285
1986	2555	330	1700		125	400
1987	2585	320	1700	40	125	400
1988	3217	1226	1400	66	140	385

*Stockpile was completely worked off.

Interest is increasing in scandium and its compounds. Its widest application is in high-power metal-halogen light sources. It is also included in $Gd_3Sc_2Ga_3O_{12}$, the basis of powerful lasers used in industry, communications networks, and antimissile devices. An important new direction for scandium use is in the production of light, high-strength scandium-aluminum alloys.

Scandium does not form separate ores, but rather is dispersed in several types of ores of other metals. Its concentration in these ores does not exceed tens, or at the best, hundreds of grams per ton. Thus, its extraction must be connected with the production of other metals.

Early production of scandium in the USSR was from uranium phosphorites. As uranium production decreased, a new technology was developed and applied industrially: extraction of scandium from hydrolytic sulfuric acid formed in the production of titanium dioxide from ilmenites. Another technology that has been developed but is not yet in industrial practice is the extraction of scandium from scandium-vanadium ore, which contains up to 200 grams of scandium per ton of ore. Methods are being developed for extraction of scandium from magnesium-titanium ores. The reserves of scandium in such ores amount to millions of tons.

The primary feature of scandium technology is the extraction of the metal from solutions resulting from the dissolution of ores in mineral acids (Table 4). The extraction is done by organic mixtures based on diethylhexylphosphoric acid and concentrates the target component by a hundred times. The resulting primary concentrate usually contains 1.5-3.5% scandium and, depending on the source material composition, 5-50% TiO_2, 2-7% ZrO, or 0.2-10% REO.

TABLE 4. Composition of Scandium-Containing Solutions (units of g/l)

HNO_3	H_2SO_4	Sc	REO	P_2O_5	Fe^{+3}	Al	Ti
150-200	-	0.06-0.10	5-8	50-70	8-12	-	-
150-150	-	0.03-0.08	10-20	50-70	8-12	-	-
-	10	0.02-0.02	0.8	80-100	3-5	-	-
5	150	0.003-0.01	-	-	8-10	-	-
-	170-220	0.007-0.025	-	-	0.5-0.8	1.5-1.8	4.5-12.0
8-100	-	0.02-0.025	-	-	10-12	10-15	-
5	100-200	0.02-0.03	-	-	5-10	-	1.0-2.0
100*	-	0.001-0.002	0.1	-	5-10	10	2.0

*Solution after nitric acid processing of loparite tails [6].

Then the primary concentrate is dissolved in the nitric acid and scandium is extracted from the resulting solutions by mixtures of neutral organophosphate compounds, which efficiently separate the scandium from accompanying elements (REE, titanium, and others). Different combinations of the components of the organic mixtures produce scandium oxides of different purities: 99.0%, 99.9%, or 99.99%.

Scandium fluoride is an important intermediate product in scandium refining. It is a source material for production of scandium metal and scandium-aluminum alloys, as well as itself a component of new optical materials. Gaseous hydrofluorination if one of the most effective methods of scandium fluoride production. The reaction is

$$Sc_2O_3 + 6\,HF = 2\,ScF_3 + 3\,H_2O \tag{2}$$

In industrial scale, this method ensures a 99% conversion of scandium oxides and gives a fluoride of 99.9% purity.

Scandium metal is produced from scandium fluoride by reduction with calcium metal.

$$ScF_3 + 1.5\,Ca = Sc + 1.5\,CaF_2 + 41.25\text{ kcal} \tag{3}$$

This process gives "rough" scandium metal of 96% purity. Vacuum distillation then gives metal of up to 99.99% purity (Table 5). Reduction with aluminum metal gives aluminum ligatures (2-20% scandium), which are then used to produce aluminum alloys for different applications.

The economics of scandium production require that the scandium be removed from the processing solutions. Reworking of tailings, such as those at Sillamäe, would require a new processing line that would add significant expense beyond the normal costs of scandium recovery. Recent prices for scandium in the United States and Russia are given in Table 6.

The practical large-scale application of scandium in industry (aircraft, automobile, and others) would be possible at a price of USD 750-1000/kg for scandium metal in ligatures. At an optimum combination of source material and processing technology, it may be possible to achieve such prices.

TABLE 7. Impurities in 99.99% Scandium Metal [7]

Element	ppm	Element	ppm	Element	ppm
H	2	Zn	<0.01	Pr	0.1
Li	<0.006	Ga	<0.07	Nd	0.7
Be	<0.003	Ga	<-.01	Sm	<0.4
B	<0.004	As	0.6	Eu	<0.1
C	40	Se	<0.1	Gd	<0.4
N	33	Br	<0.1	Tb	0.2
O	470	Rb	<0.03	Dy	0.7
F	20	Sr	<0.02	Ho	0.2
Na	<0.2	Y	3	Er	1
Mg	<0.4	Ar	<0.1	Tm	0.1
Al	1	Nb	<0.02	Yb	<0.3
Si	0.7	Mo	<0.1	Lu	0.3
P	0.1	Ru	<0.1	Hf	<0.2
S	0.8	Rh	<0.04	Ta	2
Cl	0.9	Pd	<0.5	W	<0.5
K	0.5	Ag	<0.7	Re	<0.2
Ca	0.6	Cd	<0.2	Os	<0.4
Sc	Matrix	In	<0.06	Ir	<0.3
Ti	0.6	Sn	0.2	Pt	<0.5
V	<0.08	Sb	<0.2	Au	<0.2
Cr	4	Te	<0.3	Hg	<0.4
Mn	2	I	<0.1	Tl	<0.2
Fe	9	Ca	<0.04	Pb	4
Co	<0.1	Ba	0.1	Bi	<0.1
Ni	7	La	0.2	Th	1
Cu	7	Ce	0.4	U	3

TABLE 6. 1997 Cost of Scandium, in USD/kg

	United States	Russia
Scandium oxide, 99.0%	1400	300-350
Scandium oxide, 99.9%	2900	500
Scandium oxide, 99.9%	4400	800-1000
Scandium metal, 99.99%	172,000	6000-8000
Scandium metal, in 2% Sc-Al ligature	-	1500-2000

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INDOOR OCCUPATIONAL EXPOSURE TO RADIATION AT THE SILMET PLANT IN ESTONIA

A preliminary assessment

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1. Introduction

At the meeting of the Sillamäe International Expert Reference Group (SIERG) in Stockholm, in April 1998, a study on workers' exposure to ionizing radiation at the Silmet Plant was proposed. The Silmet Plant also wanted to include analyses of chemical components in the study. The proposal was accepted by the SIERG, and a three-person steering group was established to co-ordinate the work, one representative each from Estonia, Finland and Sweden. The steering group decided in May 1998 to separate radiological and chemical measurements from each other, because their performance would need completely different expertise. It was decided that the Finnish Radiation and Nuclear Safety Authority (STUK) and the Estonian Radiation Protection Centre (EKK), together with the Silmet Plant, would perform the assessment of occupational exposure to radiation, and the Swedish Radiation Protection Institute (SSI) would search for experts to perform measurements of chemical components. This paper deals with measurements of exposure to radiation, made in May 1998 at the Silmet Plant, and with the preliminary assessments of workers' exposure to different components of ionizing radiation.

The proposal for assessment of occupational exposure to radiation at the Silmet Plant was originally presented in 1994 by the Estonian-Nordic expert group dealing with the project on the content and the environmental impact from the waste depository in Sillamäe [1]. That project discussed the possiblity of occupational exposure to radiation when raw materials with elevated concentrations of thorium and uranium were processed at the plant. At present, two different raw materials are processed: pre-processed chlorides of rare earth metals from Solikamsk in Russia, and columbite (a mixture of minerals containing niobium) from different countries in Africa.

2. Material and methods

The measurement mission to the Silmet Plant was made in May 1998 by three experts from EKK and two experts from STUK. At the time of the mission, only one of the two processing lines was in operation, processing chloride-based raw material from Russia. Thus measurements on the other line were not possible, except for a few dose rate measurements inside the processing building.

Samples of raw material and resulting wastes were taken for gamma-spectrometric measurements in the laboratories of STUK and EKK.

Samples of airborne particulate radioactivity were collected by an air sampler (flow rate 53 m^3h^{-1}) on glass fibre filters at two working sites inside the processing building (two samples per site at different times) for later gamma-spectrometric measurements. One air sample was collected out of doors in the vicinity of the processing building. Dose rates were measured in the same places by portable dose rate meters. Dose rates were also measured in different working places in the unloading station of railway wagons transporting raw materials to the plant.

Airborne radon concentrations were measured at four sites in the processing building by taking grab samples with ZnS(Ag) scintillation cells [2]. The outdoor radon concentration was measured at one site near the processing building. Airborne thoron concentrations were measured at two working sites with the two filter method [3] (flow rate 24 l min^{-1}). In connection with the higher thoron concentration, the concentration of ^{212}Pb was also measured in order to determine airborne alpha energy concentration of the thoron daughters.

3. Results

3.1. RAW MATERIAL

According to information provided by representatives of the plant, about 2000 tons per year of the chloride-based raw material imported from Russia is processed at present. This corresponds to about 20 % of the processing capacity of the plant. ^{232}Th concentration in the raw material was estimated to be about 20,000 Bq kg^{-1} based on one measured sample. ^{238}U concentration was 4,000 Bq kg^{-1} and ^{226}Ra concentration 2,000 Bq kg^{-1}.

3.2. WASTES

Two types of waste were collected from the processing line: the final waste after processing the chlorides of rare earth metals, and the waste accumulated in the process filters (mainly $BaSO_4$). The wet process waste contained large amounts of uranium, radium, and especially of thorium. The concentrations of ^{232}Th, ^{238}U and ^{226}Ra in a wet sample were about 400,000 Bq kg^{-1}, 87,000 Bq kg^{-1} and 45,000 Bq kg^{-1}, respectively. The waste accumulated in filters contained lower amounts of radioactivity, the concentrations of ^{232}Th, ^{238}U and ^{226}Ra being about 80,000 Bq kg^{-1}, 11,000 Bq kg^{-1} and 8,000 Bq kg^{-1}, respectively.

3.3. AIRBORNE PARTICULATE RADIOACTIVITY

Airborne particulate radioactivity was measured at two work sites on the second floor of the processing building (building number 55). The first site (Workplace 1) was near where the raw material is fed into the process. Two samples of airborne aerosols were collected and combined into one sample for the gamma-spectrometric measurement. The other site (Workplace 2) was near the place where process waste (mainly $BaSO_4$) is detached from the filters. Two samples were collected at this site and measured separately. One sample was collected from outdoor air near building number 55 (Outdoors).

Concentrations of ^{232}Th are estimated from the results of ^{228}Ac and ^{224}Ra, which were nearly in radioactive equilibrium in the samples. ^{232}Th was assumed to be in the same concentration as ^{228}Ac and ^{224}Ra. The uncertainty of thorium concentrations is roughly estimated to 20%. ^{238}U concentrations were not possible to measure directly from the weak gamma peak of ^{234}Pa (1001 keV) so it was estimated on the basis of the results of short-lived radon daughters (given ^{226}Ra) and ^{235}U. The error in the uranium estimations is greater than that in thorium. The airborne particulate thorium and uranium concentrations are given in Table 1.

TABLE 1: Estimations of airborne concentrations of thorium-232, uranium-238, radon and thoron, and external dose rates at Silmet Plant.

Measurement site	**Airborne radioactivity**				**Dose rate**
	Thorium-232	Uranium-238	Radon	Thoron	μSv h^{-1}
Workplace 1	3.3 mBq m^{-3}	1.7 mBq m^{-3}	130 Bq m^{-3}	1 000 Bq m^{-3}	1.0
Workplace 2	16 mBq m^{-3}	3.0 mBq m^{-3}	230 Bq m^{-3}	7 400 Bq m^{-3}	4 - 5
Outdoors	0.5 mBq m^{-3}	0.1 mBq m^{-3}	< 20 Bq m^{-3}	-	0.16

3.4. RADON

Radon concentrations on the second floor of building number 55 varied between 40 and 230 Bq m^{-3}. The mean radon concentration was estimated to be 130 Bq m^{-3}. The measured radon concentrations at Workplaces 1 and 2 and in outdoor air are shown in Table 1. The uncertainty of individual results is estimated to be about 10%.

3.5. INDOOR THORON

Airborne thoron concentrations were measured at Workplaces 1 and 2. The results are shown in Table 1. The uncertainty of the results is estimated to be about 10%. At Workplace 2, the thoron concentration was high enough that the corresponding ^{212}Pb concentration could be measured. The result was 90 Bq m^{-3}, giving an equilibrium factor of about 0.012. This equilibrium factor is used in this report.

3.6. DOSE RATE

Measurements on external dose rate in the processing building varied between 0.5 and 5 μSv h^{-1}. Dose rates in Workplaces 1 and 2 and outdoors are shown in Table 1. Dose rate measurements were also made at the unloading station for railway wagons. The background dose rate around the unloading area was at natural background levels, varying between 0.1 and 0.15 μSv h^{-1}. Dose rates in the vicinity of the containers full of the raw material were higher, varying from about 1 μSv h^{-1} to 7 μSv h^{-1} (at the container surface). The dose rate in the cabin of the crane operator was 0.19 μSv h^{-1}. In the cabin of the truck driver, the dose rate was 2 μSv h^{-1} when the track was loaded with two containers.

External dose rates in the other processing building (processing columbite) were at the same levels as in building number 55. Other measurements in the columbite line were not possible during the mission.

4. Dose Assessments

On the basis of these few measurements, it is possible to make only rough assessments of the exposure to radiation. Information on annual working hours in different work places was not available, so estimates of annual radiation doses can not be done. Instead, estimates can be made of the working hours within which an effective dose commitment of 1 mSv will be received. Workers receiving an effective dose of 1 mSv per year incurred at work from practices are regarded as exposed workers in the Basic Safety Standards Directive of the European Union (European BSS) [4]. The European

BSS also stipulates that if an occupational exposure exceeds the effective dose of 6 mSv per year, individual monitoring shall be systematic (workers of Category A).

Dose assessments have been done for Workplaces 1 and 2, in building number 55. The assessments are based on the exposure data given in Table 1 and on the effective dose coefficient given in the European BSS [4]. The decay chains of ^{232}Th and ^{238}U in inhaleable particles are assumed to be in radioactive equilibrium with ^{224}Ra and ^{226}Ra, respectively, and the mean aerodynamic diameter of particles is assumed to be one µm. In the exposure to radon daughters, a dose/exposure coefficient of 3.1×10^{-9} Sv per Bq.h.m^{-3} is used, assuming an equilibrium factor of 0.4 [1]. In exposure to thoron daughters, there is very little information about the equilibrium factor between thoron and its progeny. The dose/exposure coefficient given in the European BSS for thoron progeny is 0.5 Sv per J.h.m^{-3} which, using the equilibrium factor of 0.012 measured at Silmet Plant, gives a dose/exposure coefficient of 4.5×10^{-10} Sv per Bq.h.m^{-3}. This conversion is used in this report. The dose assessments from exposure to airborne particulate radioactivity, by assuming a mean breathing rate of 1.2 m^3 h^{-1}, and from external gamma radiation are given in Table 2. The dose estimates from exposure to radon and thoron progeny are given in Table 3. The range of action levels for radon concentration in workplaces, recommended by the European Commission is 500 – 1,000 Bq m^{-3} [6]. For thoron exposure, there are no international recommendations for action levels.

TABLE 2. Assessments of exposure times to receive an effective dose of 1 mSv from exposure to airborne particulate radioactivity of thorium and uranium decay chains and from external gamma radiation at Workplaces 1 and 2 in the processing building for chloride-based raw material at the Silmet Plant.

Workplace	**Exposure time to receive 1 mSv effective dose, hours**			
	Inhaled ^{232}Th-^{224}Ra	Inhaled ^{238}U-^{226}Ra	External radiation	**All exposures**
Workplace 1	2 900	8 300	1 000	**680**
Workplace 2	600	4 700	220	**160**

TABLE 3. Assessments of annual radiation doses from exposure to radon and thoron daughters at Workplaces 1 and 2 at the Silmet Plant, assuming an annual working time of 2 000 hours.

Workplace	**Effective dose from annual working time of 2 000 hours, mSv**	
	Inhaled Rn-daughters	Inhaled Tn-daughters
Workplace 1	0.8	0.9
Workplace 2	1.4	6.7

5. Conclusions

The main pathways of indoor occupational exposure to radiation at Silmet Plant are inhaled thoron daughters, external radiation, and inhaled particulate radioactivity. The exposure time to receive 1 mSv effective dose from inhaled long-lived particulate radioactivity and from external gamma radiation is estimated at about 700 hours at Workplace 1 and about 160 hours at the Workplace 2. The conditions at Workplace 1 can be considered rather representative of average working conditions within building number 55. The results for Workplace 2 represent radiologically the most extreme conditions of discovered working places.

The results show that exposure of workers due to inhalation of long-lived radionuclides and to external gamma radiation may well exceed 1 mSv per year and, therefore, continuous monitoring of doses of workers seems to be justified.

The radon concentrations at the workplaces were enhanced, but they are below the action levels for radon at work applied in various countries (400 - 1000 Bq m^{-3}). The thoron concentrations were exceptionally high, especially at Workplace 2. There are no international recommendations for action levels for thoron, but the comparison of doses received from thoron to those from radon indicate that further attention should be paid to thoron exposure at the Plant.

It should be remembered that the measurements were made in a plant condition where only a part of the capacity was in use. When the plant is running at full capacity, workers' exposure to radiation might be higher.

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REMEDIATION OF THE FORMER URANIUM MINE RANSTAD

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1. Introduction

The open-pit uranium mine Ranstad in southwestern Sweden (see Figure 1) was developed as a part of the Swedish Nuclear Power Programme. Uranium was extracted by percolation leaching with sulfuric acid from alum shale, which has a grade of about 300 g/t [1]. The plant was operated from 1965 to 1969. A total of 1.5 Mtons of alum shale was excavated, and 215 tons of uranium were produced. The waste in form of mill tailings consisted of about 1 million m^3 containing 100 tons of uranium and 5×10^{12} Bq of radium-226. The tailings cover an area of about 250 000 m^2.

It became obvious from operations during the 1960s and from waste handling that the weathering of mill tailings was an important environmental problem. Figure 2 presents the uranium concentration in the leachate from the mill tailings. It was already clear in the 1960s that long-term effects would become unacceptable if no actions were taken to stabilize the tailings.

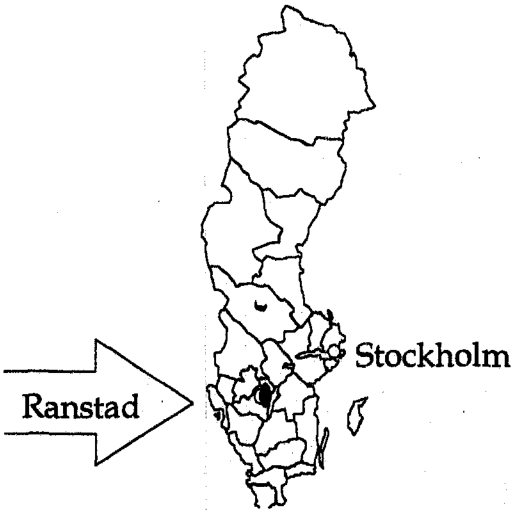

FIGURE 1. The Ranstad site

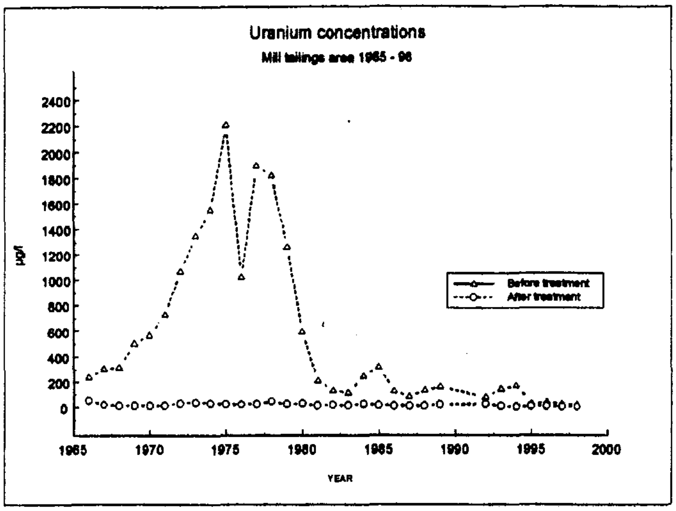

FIGURE 2. Uranium concentration in the leakage from the mill tailings area before and after treatment (1965 to 1998).

2. Laboratory and Pilot-Scale Research

Experiments and research were initiated in the early 1970s to solve the weathering problems. Weathering experiments were conducted at laboratory scale with varying mixtures of mill tailings, sludge from the neutralisation plant, and limestone. Studies were also performed to determine anaerobic conditions under water.

In the early 1970s, parallel experiments were conducted in small pits dug at the waste area. The pits were filled with mill tailings of different degree of packing, mixture, covering etc. The main results of these are [1]:

- Oxygen from the atmosphere is necessary for weathering (aerobic weathering).
- Mixing with fine-grained limestone did not stop the weathering, but the weathering products were precipitated within the mill tailings.
- Packing of the mill tailings considerably reduced the weathering.
- A cover of crushed limestone on mill tailings will, by long-term weathering, form a self-sealing layer.
- No measurable weathering was observed for mill tailings that were stored under water.

3. The Long-Term Test at the 15-kton Pile

Those early 1970s experiments resulted in a plan for a test of the dry depository method. In 1972, 15,200 tons of mill tailings (grain size < 2mm) were placed on a 80- by 55-m area. The total height of the pile was 4 m, and the slope inclination was 1:4. The mill tailings were mixed with limestone (4 percent of the total weight).

One-half of the pile was covered with a sealing layer of 0.25 m of a well-compacted moraine-bentonite mixture, and the other half was covered with crushed limestone (see Figure 3). A 1-m-thick layer of moraine and a 0.3-m-thick layer of topsoil were placed on top of both the moraine-bentonite and on the limestone. Lysimeters were emplaced at different depths for measuring the percolation of infiltrating precipitation (see Figure 3).

Results of percolation measurements from 1973 to 1981 showed that 0.2 to 3.2 percent of the precipitation percolated through the moraine-bentonite layer and and 0.3 to 4.4 percent of the precipitation percolated through the limestone layer. Measurements performed during 1986-1987 showed a precipitation percolation rate of 1.5 to 2 percent. During the same period, the flow velocity through the mill tailings was estimated to be about 3 cm per year.

No weathering was observed beneath the sealing layers on top of the test pile. No weathering was observed beneath the sealing layers on top of the test pile, while the weathering depth was 15 to 20 mm in the slope area.

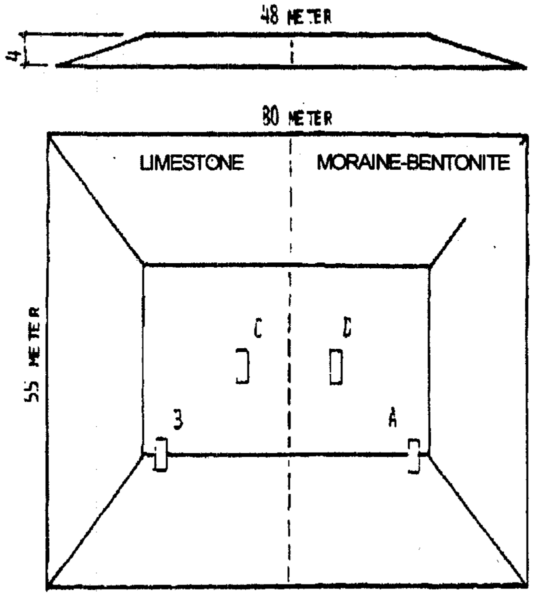

FIGURE 3. The 15-kton test pile with two types of cover material: limestone and a mixture of moraine-bentonite. Sites marked A, B, C, and D are sampling sites.

4. Remediation Techniques

Development of different remediation techniques involving both wet and dry methods was based on the results of the different studies performed at the Ranstad Site. In 1975, a depository of leaching residues within dense walls and beneath water was suggested for waste management for eventual continuation of the operation of the uranium mine.

However, when the final remediation of the Ranstad site was planned at the end of the 1980s, the dry method was chosen [2] and was accepted by the regulatory authorities in 1990.

Within the disposal area at Ranstad, a sufficient quantity of an especially favourable type of moraine was identified that contained a fine fraction of clay shale particles. This material would provide the required permeability without the addition of bentonite. A good supply of the moraine necessary for the protection layer was available both within the disposal area and in the open pit.

After careful grading and compacting of the leaching residues, the moraine for the sealing layer was spread with a laser-controlled grader and compacted with a vibrating roller. The resulting tight layer was strictly controlled by laboratory and field testing of such factors as moisture content, fine particles and compaction. Figure 4 presents a cross-section view of the depository cover system (see also TopSeal 96 [4]).

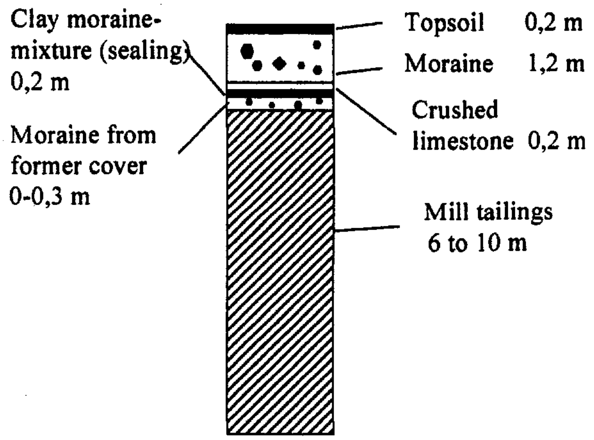

FIGURE 4. Cover system for the mill tailings depository at Ranstad Site.

5. Results of the Remediation

The goal of remediation at the Ranstad site is to reduce the effects of mining and drilling activities to levels that are acceptable in relation to the background values of the surroundings. A comprehensive monitoring programme was conducted to assess the water quality within the disposal area in the former open pit, in Lake Tranebärssjön, and in adjacent areas.

Primary restoration at the site was completed at the end of 1992 with installation of the cover system on the mill tailings. It can be concluded, five years later, that the cover system works as planned [4]. Infiltration through the sealing layer is 10 to 15 percent of precipitation. Further, oxygen diffusion through that layer is low. The concentration of radioactive elements and heavy metals in the leachate has been reduced to about one third of the concentrations before remediation (see Figure 5). Leachate from the mill tailings still must be treated

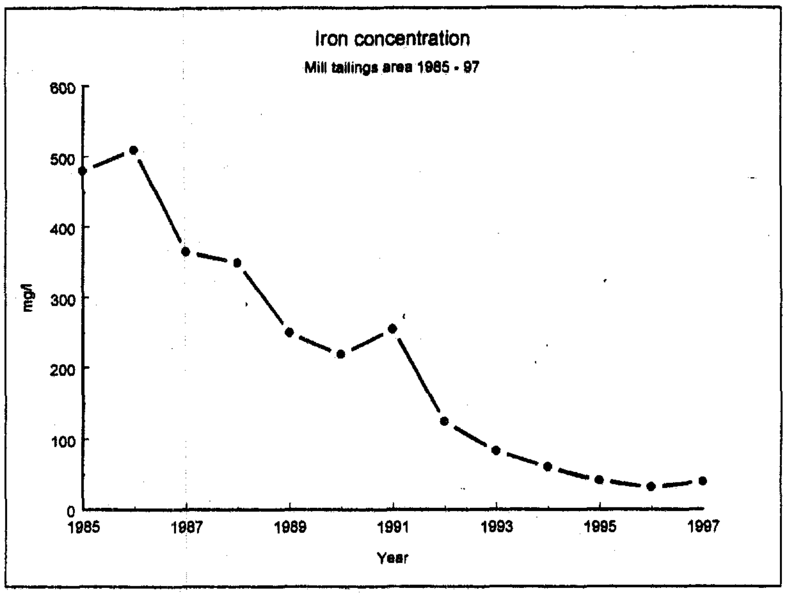

FIGURE 5. Concentration of iron in the leachate from the mill tailings area 1985 to 1997

before it can be released to the natural environment. However, the quality of this treated water meets the regulatory requirements for the site.

6. Conclusions

More than twenty years of experiments, tests, and development of remediation techniques have resulted in the remediation of the former uranium mine Ranstad. Five years after sealing the mill tailings, contaminants in the water have been reduced to about one third of the concentrations that were present before the remediation. Therefore, the sealing layer works as planned to minimise infiltration of precipitation and diffusion of oxygen. Furthermore, the water quality, after treatment, fulfils regulatory requirements.

However, the pollution levels in the former open-pit mine are still high compared to the background values. It will take long time to wash out the weathering products from the surroundings of the recent lake area.

7. References

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3. Stiglund, Y. et al (1998). The remediation in Ranstad. Release and Recipient Monitoring in 1997, Studsvik/ES-98/11.
4. TopSeal 96 (1996). Demonstrating the Practical Achievements of Nuclear Waste Management and Disposal. International Topical Meeting. Organised by the European Nuclear Society in Collaboration with the Swedish Nuclear Society.

REMEDIATION OF TAILINGS DAMS

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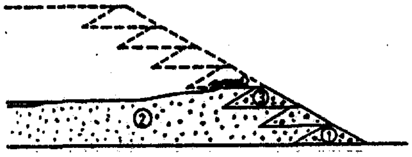
1. What Is a Tailings Dam?

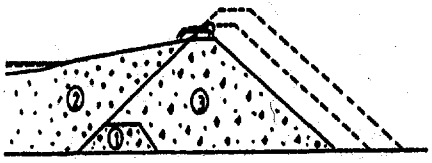
Tailings are a waste product of mining activities. The ore is crushed in processing to a size less than 0.1 to 1.0 mm, the metal content is removed, and the remaining product is called tailings. Normally, the tailings will be pumped as slurry to a sedimentation pond that is surrounded by dams and, sometimes, natural heights that form a tailings dam. Tailings dams contain both solids and water that make them similar to conventional water dams. However, their design life must be significantly longer as waste repositories because of the long-lived nature of the radiological material contained within them.

Tailings dams are typically constructed through continuous deposition. This deposition can occur by three different methods: the upstream, the centreline, and the downstream methods (see Figure 1). All three methods use a starter wall and then support fill as the height of the dam increases.



Upstream

Centreline



Downstream

FIGURE 1. Three methods of raising a tailings dam and components: ① starter wall, ② deposited tailings, and ③ support fill.

2. Environmental Aspects of Tailings Dams

Environmental effects from mining activities occur in all phases, beginning with exploration, then creation of pits and waste dumps, and finally processing of ore and handling of tailings. Three items need consideration when addressing environmental problems in connection with tailings dams:

- Environmental safety.
- Land productivity.
- Aesthetics.

2.1. ENVIRONMENTAL SAFETY

The influence on environmental safety is dominated by two main factors: (1) the physical stability of the dam and (2) the safe containment of any toxic substance. However, a physically safe dam can still pollute the environment by seepage and erosion. A tailings dam must, therefore, ensure physical, radioactive, and chemical safety for both the environment and the public during operation and after closure. The release to the environment changes during the different phases of a tailings dam (see Figure 2).

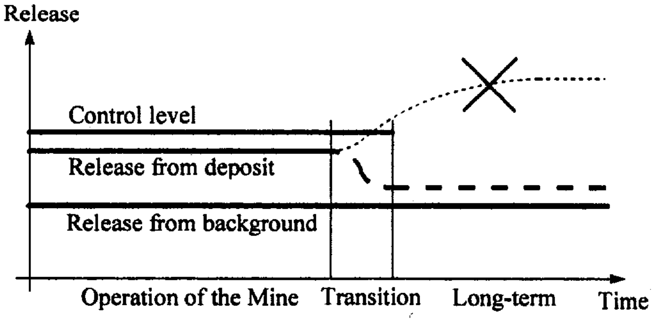
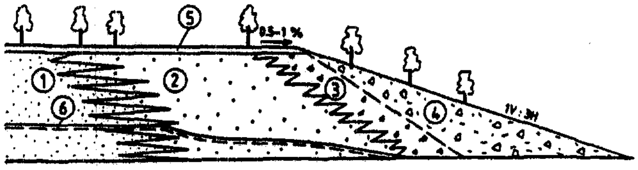


FIGURE 2. Release levels from a tailings deposit.

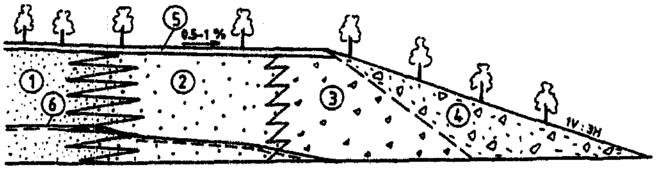
Keeping the release levels and the environmental effects below control levels during operation is normally not too difficult because the mining company is operating and maintaining the dam. During the transition phase, measures have to be taken to reduce the release level to a level close to background levels. This low release level then has to be kept low without any additional measures during the long-term phase, which in Sweden is considered to be a period of more than 1,000 years or 10,000 years, if possible. How can we ensure that a remediated tailings dam will be environmentally safe for 1,000 years? As there is no such experience with tailings dams, we have to find the answers elsewhere.

2.1.1. *Remediation Methods*

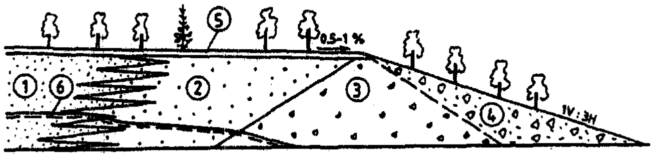
Dewatering the deposit and underwater disposal, or a combination of the two, are the two methods available. Underwater disposal means the tailings will be covered with water by creating a lake on top of them. This method is much more than dewatering and can cause a more dramatic and harmful effect on the environment if a failure occurs. Therefore, we do not accept underwater disposal in Sweden for tailings dams containing radioactive wastes. Tailings dams constructed in Sweden have to be dewatered and covered by suitable materials, such as soil and membranes, to prevent leakage of any toxin and also to prevent infiltration of water. See Figure 3 for examples of remediated dewatered deposits.



Upstream dewatered deposit



Centerline dewatered deposit



Downstream dewatered deposit

FIGURE 3. Dams for dewatered deposits. ① fine tailings, ② coarse tailings, ③ support fill, ④ support fill, long-term stable, ⑤ impervious cover and erosion protection, and ⑥ phreatic surface.

2.1.2. *Failure Mechanisms*

Irrespective of the method used, three fundamental failure mechanisms must be considered to ensure physical stability and adequate containment of the radiologically contaminated material. These three mechanisms are

- Long-term geotechnical stability.
- Extreme events.
- Slow deterioration.

Long-Term Stability. For dewatered deposits, stability is usually not a big problem because of the low phreatic surface. It is desirable to have a phreatic surface low enough to be considered ground-water pressure instead of excess water pressure.

Extreme Events. Extreme events are also not as critical for dewatered deposits as for underwater disposal. Liquefaction from an earthquake is unlikely if the deposit is permanently unsaturated. To avoid floods, diversion trenches may be required if the drainage area is large.

Slow Deterioration. Slow deterioration can damage the dam. Experiences and studies of natural formations similar to tailings dams indicate that a slope flatter than 1V:3H is often reasonably stable. This flatter slope prevents erosion by water and wind and also supports vegetation. Erosion, frost, weathering, vegetation, and animal intrusion must be considered and require a good cover material of natural origin, such as rock and soils. Natural formations with properties similar to those used in tailings dams have been in existence since the last Ice Age (approximately 7,000–15,000 years ago). A typical Swedish moraine exposed to chemical weathering since the Ice Age is unaffected below a depth of approximately 40 cm (see Figure 4). These studies support the use of natural materials to meet the longevity requirements necessary to stabilize long-lived radioactive waste.

Today's artificial materials, such as concrete, plastic, and asphalt, have a lifetime of 100 years or less, which means artificial materials can only give a temporary solution. Therefore, artificial materials must be avoided if possible.

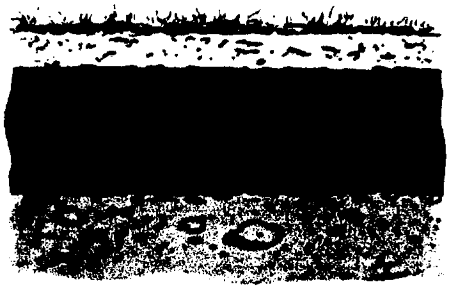


FIGURE 4. Glacial moraine exposed to chemical weathering since the Ice Age (7,000–15,000 years ago).

2.2. LAND PRODUCTIVITY AND AESTHETICS

There are still two aspects to consider after the tailings dam is environmentally safe: land productivity and aesthetics.

When a deposit is no longer in use, it must be remediated to form a natural part of the landscape. Dams should be contoured aesthetically, and the area should be brought back to the same level of productive use that was present prior to mining activities.

Aspects that should be considered with respect to aesthetics include, but are not limited to, wildlife, agriculture, forestry, and recreation. Figure 5 presents an illustration of an underwater disposal method used to create a lake that naturally fits into the landscape.

FIGURE 5. Artistic impression of underwater tailings deposit.

REMEDIATION CONCEPTS FOR URANIUM MINING OPERATIONS IN CEEC

A Phare Multi-Country Environmental Sector Programme

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Karuweeg BV
Netherlands

1. Introduction

Several Central and Eastern European countries (CEEC) have a history of uranium mining and milling. The uranium was partly used for power generation and partly for military purposes by the former USSR. The facilities were mostly operated without sufficient regard to economics and often without appropriate care for the environmental and health consequences. The nature of the environmental impact depends on the kind of activities, but typically originates from open pits, underground mines, ore and waste rock dumps, heap leaching operations, tailing ponds and processing plants. Most of these liabilities have become a source of contamination. Either by radionuclides (daughter products of uranium decay), heavy metals (other heavy metals are often associated with uranium), or chemical agents, such as acids used in uranium ore mining or processing. The contamination of the surface water, ground water and especially of potable water sources is of particular concern. All CEEC concerned wish to improve this situation.

Due to the relatively low grades of the uranium ore deposits in CEEC and due to the steady decrease of the uranium market prices, uranium production is no longer deemed economic in most of the CEEC. Consequently, the mining and milling activities have either been terminated or drastically reduced in volume. Governments of countries with current or former uranium mining and milling industries will have to provide means and funds to remediate the existing liabilities. Due to the existing economic situation within the nine CEEC facing these problems, foreign technical and financial assistance is essential.

2. The Phare MCE Uranium Remediation Programme

The European Commission (EC) has initiated this Phare Multi-Country Environmental Sector programme "Remediation Concepts for the Uranium Mining Operations in CEEC" in order to develop appropriate management practices for these liabilities and to assist the governments of these CEEC in planning rehabilitation work of the sites concerned.

This programme started in July 1996 and it consists of two different parts.

The first part, Part A, has produced a complete inventory of all uranium liabilities in the nine CEEC involved. These environmental liabilities have been categorized and the environmental impact of these categories has been assessed in a preliminary way. In addition, the legal frameworks regarding licensing of this remediation as well as the regulations concerning waste water releases have been reviewed for all the countries involved, in comparison with the situation in EU and other Western countries.

The second part, Part B, consists of a series of Pilot Projects. Each of these pilot projects will take care of technical assistance for a specific topic in the remediation of these environmental liabilties. In doing so, the project will cover both the common aspects of such a topic for all CEEC involved and the specific aspects of a field application of the assistance to one of the sites in one of the countries.

3. Objectives

The overall objectives of this programme are threefold:

- to inventory the existing situation,
- to implement pilot projects, and
- to support the co-operation between the involved countries.

The objectives of the different programme components, especially those of the pilot projects, are much more structured.

4. The Pilot Projects

The generic part A of the programme was completed in August 1998. Since May 1998 the second part of the programme is running. It consists of seven different pilot projects. Three of these projects deal with different aspects of tailing pond remediation.

These topics are:

- stabilization of the fines in the tailing ponds interior, in order to be able to carry loads and to be capped
- development of the water balance of a tailing pond, during and after remediation
- reshaping and covering a tailing pond, including its effect on dam stability

Other topics for pilot projects are:

- development of overall water management systems for large and complicated areas with extensive uranium mining and milling
- closure and flooding of underground uranium mines
- assessment of the effectiveness of historic remediation steps
- development of an assessment method for the numerous smaller uranium liabilities in CEEC

These pilot projects are focussing on large-scale remediation projects in Hungary, the Czech Republic, Romania, Slovakia, Albania, Estonia, and Bulgaria. Three of these pilot projects have started. The other four are expected to start in autumn 1998.

Most of these pilot projects are connected to one another. Therefore extensive communication lines are scheduled, in order to disseminate the experiences and results to all possibly interested parties in CEEC. This communication has been secured by an extensive project guidance not only by the host country, but also by other interested CEEC. In addition, all reports will be distributed among the supporting countries for each pilot project. Finally, for each pilot project one or two Experts Workshops are scheduled, for optimum exchange of information, experience and results.

A first Experts Workshop on "developing the water balance of a tailing pond under and after remediation" took place in Czech Republic, September 26, 1998. A second Experts Workshop on stabilizing tailing pond interiors is scheduled for October 13 and 14, 1998, in Hungary. A third Experts Workshop on closure and flooding of deep underground uranium mines is scheduled in Romania, March 18, 1999.

More specific information about the different pilot projects is presented in the following sections.

4.1 PILOT PROJECT NR. 1: TECHNICAL PLANNING OF LONG-TERM STABILITY OF TAILINGS POND INTERIORS

4.1.1 Project Objectives

The objectives of this pilot project are:

- To develop specific criteria and to identify key parameters for a systematic approach to the stabilization of the sludge interior of uranium tailing ponds that can be applied to the remediation of all tailing ponds in CEEC.
- To apply this approach to a specific site at Pecs, as a practical learning case.

4.1.2 The General Problem

Uranium tailing ponds usually have large surfaces through which they collect a lot of water from precipitation. Part of this water will seep through the dams into the surface water or even through the bottom layers into the underground. This can lead to the pollution of underground (drinking) waters and even to geotechnical instability of the pond. Therefore remediation always looks for the possibility to minimize water from precipitation entering the pond's interior.

This interior consists of very fine grained sludges, with a high pore water retaining capability. This results in very weak material, hardly capable to carry loads. Minimizing water will require reshaping the surface of the pond to improve the rainwater run-off and covering the pond's surface with low permeability layers to minimize this water entering the pond's interior. In order to realize this, the mechanical stability of the pond's interior has to be improved so that it can carry heavy equipment to do the reshaping and covering and that its new shape will be long-term stable.

This situation is similar for all other uranium tailing ponds in CEEC.

4.1.3 The Tailings Ponds at Pecs

The two tailing ponds at Pecs contain 20 million m^3 of sludge, covering 150 hectares. These ponds were built without a good isolating layer to the underground. Due to this, polluted water is constantly leaking from the ponds into the underground. This underground is part of the water reservoir used to supply drinking water to the city of Pecs. Local pumping depresses the local water table. The pumped water is decontaminated in the processing plant's water treatment facility. This processing plant will be terminated at the end of 1997. In 1998 preparations for the remediation of the tailing ponds started.

Considering the urgency of the solution at Pecs and considering the time schedule involved, the tailing ponds at Pecs were considered to be very interesting for a case application of the systematic approach for stabilizing tailing pond interiors.

4.1.4 Further Applicability

Hungary needs this project to be done as one of the basic elements for detailed remediation planning and design. Six other CEE countries possess another 27 uranium tailing ponds. All these ponds need improvement of the stability of their pond's interiors for different reasons.

4.1.5 Project Management Details

Submitted by : MEV, Pecs, Hungary
Supported by : Bulgaria, Czech Republic, Estonia, Poland, Romania, Slovenia
Duration : 1 year
Total costs : 20.000.000 ECU (total remediation costs for the tailing ponds)
Phare part : 450.000 ECU (this pilot project only)
Project : started May 1998

4.2 PILOT PROJECT NR. 2: PREDICTION OF THE DEVELOPMENT IN TIME OF THE WATER BALANCE OF A TAILINGS POND

4.2.1 Project Objectives

The objectives of this pilot project are:

- The development of a systematic approach of how to design an affordable and long term stable water management system of (remediated) tailing ponds
- The application of the results to a specific site at Rozna, as a practical learning case.

4.2.2 The General Problem

All uranium tailing ponds are part of ore milling and processing plants. The ponds collect the residues after milling and leaching has taken place. These residues are pumped into the ponds with excess water. This excess water is returned from the pond to the water treatment facility of the processing plant. Depending on the processes used in the processing plant, the chemistry of the tailing pond can vary widely, even within one pond.

As long as the processing plant is operating, its water treatment facility will take care of the excess water, including the water from precipitation, groundwater leaking into the pond and pond water seeping out through the dams or even through the bottom of the pond. This will establish a water balance that is more or less under control.

As soon as the processing plant is no longer operating, the water balance changes, even more in the case of ungoing or completed remediation. The amounts and the chemistry of the water that needs treatment will change drastically with time. Usually, the original water treatment facility of the original processing plant is no longer suitable. Therefore a new water treatment facility will be needed and it will have to be designed for the long future, capable of taking care of all variations in quantities and of the changing chemistry.

This situation is similar for all tailing ponds related to uranium ore processing.

4.2.3 The Rozna Tailings Pond

The processing plant at Rozna has two tailing ponds, a large one (9 million m^3 deposited sludge) and a small one (0.9 million m^3). At present the processing plant is still working, taking care of the treatment of 700,000 m^3 of excess water each year.

It is expected that the production of ore will be stopped soon. For 1998 it will be decreased to less than 50 per cent of the previous production volume. The tailing ponds are situated in a drinking water protected area between Prague and Brno.

Considering the expected changes in the water balance of the Rozna ponds, their strict constraints as well as the time schedule involved, these tailing ponds at Rozna are very interesting for a case application of the systematic approach to be developed.

4.2.4 Further Applicability

The Czech Republic contains 12 other tailing ponds. All of these still need remediation. Also, all supporting countries have tailing ponds, most of them with similar needs for water balance management. The total number of tailing ponds in these countries is 29.

4.2.5 Project Management Details

Submitted by : DIAMO, Straz pod Ralskem, Czech Republic
Supported by : Bulgaria, Estonia, Hungary, Poland, Romania, Slovenia
Duration : 1 year
Total costs : 780.000 ECU (only this part of remediation preparation)
Phare part : 390.000 ECU (50 per cent)
Project : started May 1998

4.3 PILOT PROJECT NR. 3: TECHNICAL PLANNING OF UNDERGROUND MINE REHABILITATION

4.3.1 Project Objectives

The objectives of this pilot project are:

- To develop a methodology for planning the best way to close an uranium mine, to flood it, and to minimize the environmental consequences.
- To apply this methodology to the Ciudanovita mine, as a practical learning case.

4.3.2 The General Problem

Underground mining of uranium mostly takes place under the groundwater table. During mining activities, this groundwater table is depressed locally by pumping. Usually these so-called mine waters have to be decontaminated before they can be released into surface waters. Due to this underground mining of uranium, a large underground volume of rock is removed. Depending on the kind of mining techniques applied, part of the underground structure may be cracked or disturbed and thereby more easily accessible for water. The result of the mining is a large underground surface, in connection with remaining uranium ore bodies (poor concentrations, or sometimes even rich), and the structure of that surface is rather permeable for water. When mining ends, the pumping is stopped and the locally depressed groundwater table is restored. This water will contact the uranium left in the mine system. This will lead to enhanced concentrations of uranium and daughter elements in this mine flooding water. After some time this local water will mix with other underground and surface waters surrounding the site and will start spreading out. In order to avoid high concentrations of unwanted (radio)chemicals in ground and surface waters, for a number of years some kind of water depression by selective pumping will remain necessary, such in combination with a purification plant for the water pumped out. Key questions are for how long this pumping has to be done, how much water has to be pumped, from what depth and what will be the needs for water decontamination.

This set of questions is similar for all mines that have to be flooded in the CEEC.

4.3.3 The Mine at Ciudanovita

The mine at Ciudanovita is a typical uranium mine, with a complicated internal structure and different pumping levels. Therefore, several options for flooding the mine can be analysed for their environmental impact and their economic consequences. Considering the actual status of the Ciudanovita mine, the variety of options for flooding and the decision of the Romanian government to close it, this mine is quite interesting for a case application of a methodology developed for optimizing the close-out of uranium mines.

4.3.4 Further Applicability

Romania counts 128 shafts and adits, used or still in use for uranium mining. Some of these are in the underground water regime. In addition, the other eight CEE countries count another 3,503 shafts and adits, of which by far the greatest number still have to be flooded. Therefore, the potential of the further use of a systematic approach for mine flooding is large.

4.1.5 Project Management Details

Submitted by : Rare Metals Autonomous Regie, Bucharest, Romania
Supported by : Bulgaria, Czech Republic, Hungary, Poland, Slovakia, Slovenia
Duration : 18 months
Total costs : 3,400,000 ECU (estimated total remediation costs for the mine Ciudanovita)
Phare part : 450,000 ECU (this pilot project only)
Project : started September 1998

4.4 PILOT PROJECT NR. 4: TECHNICAL PLANNING OF TAILING POND REMEDIATION, INCLUDING LONG-TERM DAM STABILITY

4.4.1 Project Objectives

The objectives of this pilot project are:

- The development of a systematic approach for reshaping and covering tailing ponds resulting from the processing of uranium ores, with special emphasis on realizing long-term dam stability.
- To apply this systematic approach as a learning case to the tailing pond at Sillamäe.

4.4.2 The General Problem

Tailing ponds from uranium ore processing plants are large reservoirs in the open air, where the residues from processing are deposited. Extraction of uranium never reaches 100 per cent; therefore the residues contain significant amounts of uranium as well as all daughter elements of the original uranium ores. Depending on the chemical processes that were used in the plant and depending of the ores that have been processed, these ponds also contain other hazardous materials.

These tailing ponds may be deposited in a local valley, with a closing dam at the bottom end of the valley, on a slope, with a dam on two or a three sides surrounding the pond, or even a ring type dam, surrounding the pond in flatter regions.

It appears that these dams have not always been constructed in a long-term stable way. A few of them already show sliding. Also seismic conditions have not always been taken into account correctly.

For remediation planning for a tailing pond, ensuring the long-term stability of the dams is necessary. Dam stability will also be affected by the overall remediation of the tailing pond, including drying-out of the pond and reshaping and covering the surface of the pond. If this stability is not good enough, a systematic approach of how to improve this stability in the most effective way, taking into account the effects of the tailing pond remediation as well, will be an useful tool for the remediation of 29 tailing ponds in seven CEE countries.

4.4.3 The Tailing Pond at Sillamäe

The tailing pond at Sillamäe is located at the shore of the Gulf of Finland. The foot of the dam is very close to the surface of the Baltic sea. The pond has been constructed on a slope, which is tilted in the direction of the sea. Measurements show that the dam of this pond is sliding slowly down this layer, into the Baltic Sea. This sea itself, at high tides in bad weather, attacks the foot of this dam by wave action. This latter could enhance the sliding of the dam. Stabilizing the dam is necessary to prevent a collaps.

Considering the actual status of the pond and the urgency for restoring the long-term stability of the dam, this tailing pond at Sillamae is a good example for a first case application of the systematic approach for overall tailing pond remediation, reshaping and cover its surface and ensuring long-term dam stability.

4.4.4 Further Applicability

Sliding risks do not occur only in Estonia. Bulgaria, the Czech Republic, Hungary, Poland, and Slovenia also have tailing ponds facing stability issues that need to be improved in connection with their remediation.

4.4.5 Project Management Details

Submitted by : RAS Silmet, Sillamäe, Estonia
Supported by : Bulgaria, Czech Republic, Hungary, Poland
Duration : 18 months
Total costs : 20,000,000 ECU (estimated total remediation costs for the tailing pond)
Phare part : 448,000 ECU (this pilot project only)
Project : scheduled to start November 1998

4.5 PILOT PROJECT NR. 5: EFFICIENCY OF FORMER REVITALIZATION AFTER URANIUM MINING

4.5.1 Project Objectives

The objectives of this pilot project are:

- To develop an appropriate methodology for the evaluation of the efficiency of historic remediation of uranium liabilities.
- To apply this methodology to a selected group of historically remediated uranium liabilities near Novoveska Huta, as a learning case.

4.5.2 The General Problem

Mining and milling of uranium have taken place for several decades in CEEC. Today, many of these sites are no longer used or not even attended. In a few cases some remediation has taken place. Most of this took place under regulations, that are not as stringent as the regulations of today. Methods used were rather simple, and many environmental aspects were not taken into account.

Until now no evaluation has been made of the effectiveness of these former remediation actions. Especially important will be the assessment of whether and to what degree radiochemicals and other harmful elements still are migrating into the environment. Also important will be the assessment whether these formerly remediated uranium liabilities meet today's (EU) standards, or where they have to be improved.

This situation occurs in nearly all CEE countries.

4.5.3 The Village Novoveska Huta

The region around the village Novoveska Huta in Slovakia contains several underground mines, waste rock dumps, dumps with unsold uranium and other issues. Especially in this part of Slovakia, where the mining industry was closed in 1989, a variety of historic remediation steps can be evaluated for their efficiency. This area is situated in the second largest national park in Slovakia.

4.5.4 Further Applicability

Most of the CEE countries have old uranium liabilities that have undergone only simple remediation steps or none at all. The development of a method for systematic evaluation of the efficiency of these historical choices will be an important tool for all countries. Historical measures might be sufficient, which will lead to a reconsidering of formerly used methods, or they might turn out to be insufficient, which would mean that such a "remediated" liability would need further remediation.

4.1.5 Project Management Details

Submitted by	: Uranpres, Spisska Nova Ves, Slovakia
Supported by	: Albania, Bulgaria, Czech Republic, Hungary, Poland, Romania, Slovenia
Duration	: 11 months
Total costs	: 230,000 ECU (only this pilot project)
Phare part	: 180,000 ECU (only this pilot project)
Project	: scheduled to start December 1998

4.6 PILOT PROJECT NR. 6: DEVELOPMENT OF A COMPREHENSIVE METHOD FOR THE IMPACT ASSESSMENT OF SMALLER URANIUM LIABILITIES AND ITS APPLICATION TO THE RADIOLOGICAL EFFECTS CREATED DURING URANIUM EXPLORATION IN ALBANIA

4.6.1 Project Objectives

The objectives of this pilot project are:

- The development of a systematic method for the assessment of the impact of the smaller uranium liabilities on environment and on human health, suitable for use in all CEE countries.
- To apply this developed method to the assessment of the impact of the uranium liabilities in Albania.

4.6.2 The General Problem

Many of the environmental liabilities resulting from uranium mining and milling are large and quite visible. Apart from these larger uranium mining and milling liabilities in CEEC, a significant group of smaller liabilities has also been identified in these CEEC. These are, for instance, the residues from exploratory mining activities. In some of these CEE countries, even some very old liabilities are present, connected to mining activities of long ago, but containing significant amounts of uranium ore and its daughter products. Some of these historic liabilities even date from before 1940.

Ignoring these smaller uranium liabilities would be a mistake, since they can contain quite a lot of uranium ore, and since they can represent a quite substantial risk. The existing full-scale impact assessment methods are too complex and too expensive to be of use for assessing these small liabilities one by one. For assessing the potential risks of these numerous smaller liabilities, a more comprehensive systematic approach is needed.

4.6.3 The Smaller Uranium Liabilities in Albania

Exploration for uranium ore bodies in Albania was carried out between 1961 and 1989. During this period 379 adits were created with a total length of nearly 37 km, and in addition 700 boreholes. The materials that were removed were disposed at the surface in 379 dumps, with a total volume of 140,000 m^3. The total area that was affected by drilling and by exploratory mining amounts to 21.25 km^2, distributed over 6 districts. In Albania, the uranium ore was deposited on the same dumps as the mine rocks: at the surface. No measures were taken to prevent its further use. In some districts, the villages start at the edge of the dumps. No remediation has taken place. Albania even lacks equipment for systematic measurement of concentrations of radionuclides in water, soil and air.

This makes Albania an interesting case for a field application of such a comprehensive impact assessment method.

4.6.4 Further Applicability

Seven other CEE countries also have large amounts of similar small uranium liabilities. The availability of such a comprehensive assessment method would be an useful instrument for remediation planning.

4.6.5 Project Management Details

Submitted by	: Ministry for Health and Environment, Albania
Supported by	: Bulgaria, Czech Republic, Hungary, Poland, Romania, Slovakia, Slovenia
Duration	: 18 months
Total costs	: 210,000 ECU (total costs for this project)
Phare part	: 185,000 ECU (88 percent of total costs)
Project	: scheduled to start December 1998

4.7 PILOT PROJECT NR. 7: MANAGEMENT AND CLEAN-UP OF GROUND AND SURFACE WATER POLLUTED WITH RADIONUCLIDES AS A RESULT OF URANIUM MINING AND PROCESSING ACTIVITIES IN THE BUHOVO AREA

4.7.1 Project Objectives

The objectives of this pilot project are:

- To create a suitable methodology for the development of an overall water management strategy, applicable to larger areas in CEE countries, affected by uranium mining and/or milling activities.
- To apply the methodology to the affected area of Buhovo, resulting in the development of an overall water management strategy for Buhovo and in providing a plan of action for the implementation of this strategy, outlining the technical measures, needed to realize the objectives.

4.7.2 The General Problem

Uranium mining in CEEC has taken place in many different ways. Open pit mining, conventional underground mining, undergound in-situ leaching, heap leaching, underground block leaching, above-ground processing of ores, and even the open air storage of ores and residues, all influence the water quality in these regions. In many countries, several of these activities were concentrated in the same regions. These mining regions can be quite large and in many cases they are situated in mountain areas with complex hydrogeology, which in its turn may be affected by the mining activities. All this can complicate proper management of water quality for those regions.

A systematic method of how to develop the best strategy for water management in such regions will be an effective remediation tool for several different areas in all nine CEE countries.

4.7.3 The Situation at Buhovo

The Buhovo mining area is situated at the Southern ridge of the Balkan mountains, at a 20-25 km distance from Sofia. This is one of the larger uranium mining and milling areas in Bulgaria, covering an area of approximately 70 km^2. Mining started more than 50 years ago and lasted until 1992. The Buhovo region has seven underground mines. More than 55 km of shafts have been driven, including seven pits and 153 horizontal mining facilities. In total, 198 dumps of mine tailings were created, with a total volume of 4 million m^3. Underground chemical leaching techniques were applied, leaving approximately 160,000 m^3 of sodium leachate in the underground, most likely due to absorption. Mines were flooded without any precaution and without any water treatment foreseen. In addition, a processing plant has been operating for many years, initially without any retention of waste waters. The plant released its waste waters directly into the river system near the plant, contaminating the river bed and the banks. At present there are two large tailing ponds, covering an area of 100 ha and containing several million m^3 of processing residues. These ponds are leaking into the underground, the dam of the pond is leaking into the river system, and the dam stability is not in compliance with seismic stability regulations.

This all makes the Buhovo region very interesting for a first application of this developed method.

4.7.4 Further Applicability

Similar areas can be found in other parts of Bulgaria and in nearly all other CEE countries.

4.7.5 Project Management Details

Submitted by : Committee for Energy, Bulgaria
Supported by : Albania, Czech Republic, Estonia, Hungary, Poland, Romania, Slovakia, Slovenia
Duration : 1 year
Total costs : 551,500 ECU (this preparation project only)
Phare part : 413,500 ECU (75 percent of this preparation project)
Project : scheduled to start January 1999

5. Future steps

The generic part of this Phare programme has been completed.

The pilot projects, meant to assist the CEE countries in organizing their national remediation programmes, thereby benefitting from Western experience in this kind of environmental remediation, will be completed Summer 2000.

At this moment, full-scale remediation programmes are under development or being implemented in most of these Central and Eastern European countries. All these programmes need substantial investments. Preliminary cost estimates for these necessary environmental investments vary from 3,000 to 5,000 million ECU. These environmental investments consist in large part of local labour. In many of these countries, the cost for labour still is very low as compared to EU labour costs. Therefore, these cost estimates will become significantly higher when the costs for labour in these countries rise to levels that are more comparable to those in Western European countries.

The European Union has made available funds to assist these CEE countries to bridge the gap between Western and Eastern Europe. For implementing this assistance both National Phare programmes and other financing instruments have been developed or are under development, especially for infrastructure and environmental problems.

For several uranium remediation problems in Phare countries that conflict with the European Union's legislation, directives or standards, such environmental investment assistance instruments might be applicable in the near future.

WISMUT EXPERIENCE IN REMEDIATION OF URANIUM MILL TAILINGS PONDS

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1. Introduction

The post-war production of uranium by the former SDAG WISMUT (a joint Soviet-German enterprise) amounted to approximately 220,000 t. After German reunification in 1990, the appraisal of the economics of the company indicated that further mining was not feasible, and production was terminated. The environmental impact of the former operations of the company identified an area of approximately 32 km^2 in the densely populated states of Saxony and Thuringia that were in need of remediation (Figure 1). To remediate the environmental hazards, WISMUT GmbH was constituted by the Federal Ministry of Economics. The remediation was estimated to require 15 years and to cost DM 13 billion.

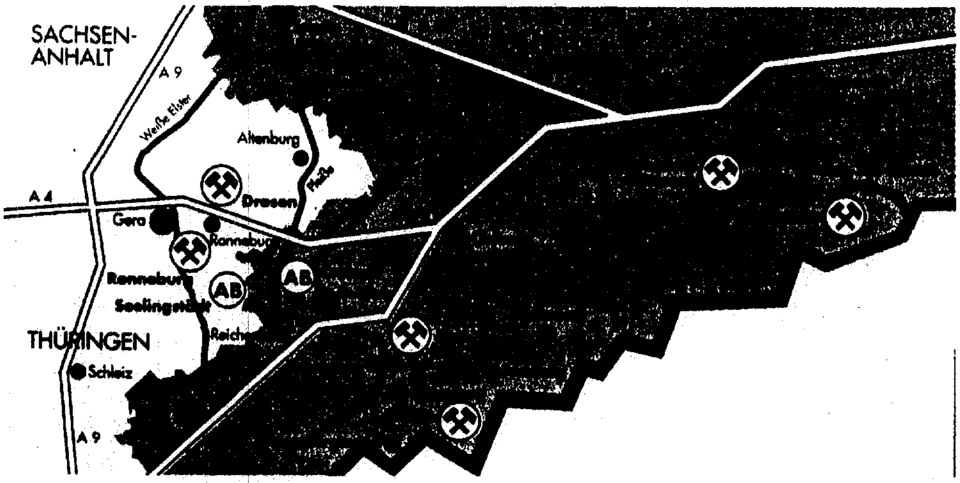

FIGURE 1. Location of the WISMUT remediation sites (AB: processing sites at Crossen and Seelingstädt).

Milling operations ceased in 1989–1990 (short after-production runs extended into 1991), leaving in place a total of 545 hectares of tailings ponds at Seelingstädt (Culmitzsch and Trünzig) and Crossen (Helmsdorf and Dänkritz) with a total tailings volume of 150 million m^3. Table 1 presents the key characteristics of the tailings ponds. In addition to the listed contaminants, the pond water at Helmsdorf contains up to 90 mg As/liter and the tailings pore water contains up to 6 mg/liter of As.

TABLE 1. Key characteristics of WISMUT tailings impoundments

Tailings Impoundment	Culmitzsch Basin A	Culmitzsch Basin B	Trünzig Basin A	Trünzig Basin B	Helmsdorf	Dänkritz I
Tailings surface area (ha)	158.1	75.8	66.8	48.1	205.3	18.9
Water cover surface area (ha)	72.7	48.3	0	36.5	139.7	12.2
Tailings volume (Mio m^3)	61.3	23.6	13	6	45	4.6
Solid mass (Mio t)	64.0	27.0	13	6	48.9	6.6
Max. tailings thickness (m)	72	63	30	N.A.	48	23
U_{nat} in solids (t)	4800	2240	1500	700	5030	1052
Ra-226 in solids (1014 Bq)	7.9	2.4	1.3	0.5	5.5	0.38
U_{nt} in pond water (mg/L)	0.14	6.29	N.A.$^{1)}$	1.0	6.54	1.70
Ra-226 in pond water (mBq/L)	191	1232	N.A.$^{1)}$	150	1300	760
U_{nat} in pore water (mg/L)	0.3 ... 3.9	1.0 ... 16.5	1.19	N.A.	2 ... 30	10 ... 85
Ra-226 in pore water (mBq/L)	... 5000	... 2300	632	N.A.	500...2000	N.A.

The goals of the tailings impoundment remediation can be summarized as (a) stabilization of the impoundment, (b) provision of hydrological control, (c) control of contaminant seepage, (d) integration of the remediated object into the landscape, and (e) minimization of the need for water treatment, surveillance, and monitoring after remediation. On the basis of a probabilistic risk assessment, it was agreed with the regulator that a reasonable remediation is best achieved by placement of a dry cover over the tailings impoundments [1, 2, 3].

2. Appraisal of the Tailings Impoundments

Both reconstruction of the historical development and investigation of present conditions proved to be necessary for the appraisal of the tailings impoundments. The historical data used in the appraisal include the description and evaluation of the preconstruction topography and major physical features; embankment construction (foundation, embankment design, and construction details); tailings discharge/deposition history (tonnage placed per year and resulting elevations of the placed tailings surface); tailings discharge practices over the years; and records of unexpected behavior. Aerial photographs provided useful information on the location of the tailings discharge points for the operating life of the facility. Records of water balance in the impoundment, size of the pond, pond water, and seepage water quality were collected and assessed.

On the basis of historical data, a site investigation plan was developed. In the pond area of fine tailings, drilling and testing are conducted from a floating platform.

On the basis of historical data, a site investigation plan was developed. In the pond area of fine tailings, drilling and testing are conducted from a floating platform. Special drilling and vacuum sampling equipment is used to obtain undisturbed samples of the loose tailings sands and soft (fluid) underconsolidated slimes for laboratory investigations. The investigations on samples include grain-size distribution and moisture content measurements, solids specific-gravity determinations, Atterberg limits, consolidation, permeability, and shear strength.

The testing of the slimes required adjustment of the standard geotechnical equipment. The field tests most commonly used were shear-vane testing for measurement of the total shear strength and piezocone testing for estimations of shear strength, excess pore pressure, and permeability at various depths.

2.1 CROSSEN

At the Crossen Site, the smaller Dänkritz I (and Dänkritz II) ponds were established in a sand pit and a gravel pit, respectively. The large Helmsdorf tailings pond was created as a valley impoundment at the head of the Oberrothenbach Creek Valley. A large capacity impoundment was constructed with one main dam (2,200 m long with a maximum height of 58 m). Drainage occurs through the alluvial deposits of the buried valleys in the foundation of the impoundment into the Oberrothenbach Creek.

The slimes zone comprises much of the tailings area in the IAA Helmsdorf now covered by water. Figure 2 shows the void ratio of two profiles in the center of the slimes zone (HW11 and HW12). The range of void ratios is significant in spite of the uniformity of material. A decreasing void ratio with depth can be observed because of the higher effective stress with increasing depth. The lower void ratio at the base of HW 12 indicates a free-draining foundation at this location.

The observed high void ratios are consistent with the low shear-strength values measured in the slimes zone. The undrained shear strength values, determined by shear vane testing, for the pond zone are

0 to 3 m	0–3 kN/m^2
3 to 10 m	0–10 kN/m^2
>10 m	> 8 kN/m^2

Oedometer measurements in the laboratory indicated a high relative settlement of up to 28% for the slimes. From the tests, a low coefficient of consolidation, $C_v = 1$ to 4×10^{-7} m^2 /s, and low vertical hydraulic conductivity, k_v, on the order of 2 to 6 $\times$ 10^{-9} m/s, were derived.

The appraisal of the Helmsdorf data suggested that, for purposes of remediation, it is advantageous to distinguish the sandy beach, slimes, and intermediate zones. Table 2 presents characteristic geotechnical data for each zone of the Helmsdorf impoundment.

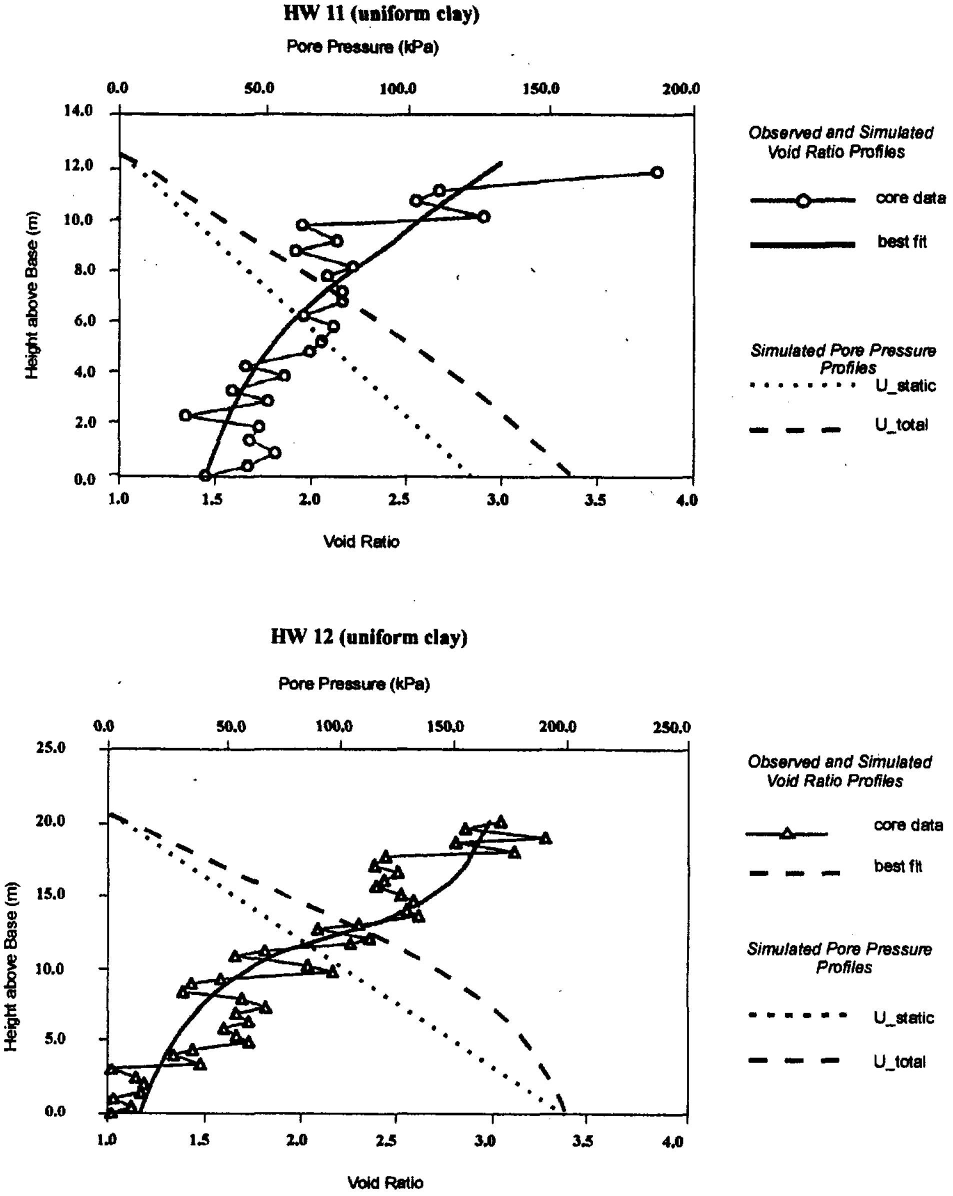

FIGURE 2. Observed and simulated void ratio and pore pressure profiles in the fine tailings of Helmsdorf for boreholes No. HW-11 (top) and HW-12 (bottom) [based on data from 4, 5].

TABLE 2. **Representative geotechnical properties of the different tailings zones in Helmsdorf [based on data from 5]**

Geotechnical Parameter	**Sandy Beach**	**Intermediate Zone**	**Slimes Zone**
Grain size			
d_{10} (mm)	0.02	0.01 – 0.002	<<0.001
d_{60} (mm)	0.1	0.08 – 0.008	0.005
d_{90} (mm)	0.25	0.1 –0.04	0.03
Average Water Content, w_n	0.10 – 0.30	0.25 – 0.65	0.45 – 0.96
Atterberg Limits			
w_P	0.15	0.15 – 0.19	0.21
w_L	0,28	0.30 – 0.47	0.54
I_P	0.14	0.16 – 0.18	0.32
Dry Density, ρ_s (g/cm^3)	2.76	2.79	2.78
Bulk Density, ρ_n (g/cm^3)	1.95	1.78 – 1.93	1.64 – 1.78
Void Ratio, e	0.55 – 1.0	0.7 – 2.0	1.5 – 3.0
Total Shear Strength, τ (kN/m^2)	20 – ≥50	5 – 50	< 5 (surface) 5 – 20 (depth)
Coefficient of Consolidation, c_v (m^2/s)	1 to $2 \cdot 10^{-6}$	$2 \cdot 10^{-6}$ to $2 \cdot 10^{-7}$	1to $4 \cdot 10^{-7}$

2.2 SEELINGSTÄDT

The Culmitzsch and Trünzig impoundments were established in worked-out open-pit uranium mines. Fractured sandstone forms the basements of the impoundments, allowing drainage from the tailings. Seepage from the impoundments is toward the Lerchenbach and Fuchsbach creeks. Settlement and piezometer measurements, as well as results of a field test, were available for the appraisal of the Culmitzsch A tailings pond.

In the field test, a 1-m-thick cover with drains at various spacings was advanced on the surface of the intermediate beach zone. Estimates of the pore water pressure were made based on piezometer measurements. The slimes, consisting of silts and clays, in this impoundment showed a moisture content of 0.7 to >1.5 and proved to be highly compressible. A coefficient of consolidation of $C_v = 1$ to 5×10^{-7} m^2/s is derived from the laboratory measurements.

The subdivision of the Culmitzsch A impoundment into zones proved to be more complex than at Helmsdorf because of the layered structure of the tailings deposit [6]. Evaluation of historical and present-day geotechnical data from Helmsdorf and Culmitzsch A lead to the conclusion that the subdivision of the impoundment into zones must be based, in addition to material characteristics, on the thickness of the tailings, discharge/depositional history, and drainage conditions. In the conceptual model of the impoundment needed for the assessment of the overall performance of the remediation, the identified tailings zones must be represented as separate elements.

3. Development of the Geotechnical Specifications for Remediation

The geotechnical specifications needed for the tailings impoundment remediation were developed from the perspective of the overall rehabilitation goals that will be achieved by capping the impoundment with an interim cover and a final cover.

The prerequisite for placement of the cap is the design of the post-remediation shape of the impoundment, which requires knowledge of the depth of tailings settlement and duration of the settlement process. By computing tailings settlement following the placement of the interim cover, the additional fill volume can be estimated and areas of placement in the impoundment identified.

The overall remediation goal of preventing long-term ponding on the surface and infiltration of water into the tailings is accomplished by placement of the final cover. For timing of the final cover placement, it is essential to estimate the completion time of the tailings settlement.

Prediction of the fine tailings settlement requires a number of steps, including matching the tailings discharge history, back calculation of the geomechanical parameters, and calibration based on actual measurements, to provide the prognoses. Only an adequately defined simulation can provide results that are sufficiently accurate to allow a meaningful calculation of the material needs for the final shape of the impoundment. Therefore, (a) no predictions should be attempted without history matching, and (b) the actual tailings behavior must be monitored and the physical model of the impoundment updated periodically for improved predictions.

3.1 DATA REQUIREMENTS

The settlement modelling of an entire impoundment with various tailings zones requires a broad set of data comprising (a) depth of the impoundment base and thickness of tailings; (b) tailings material-behavior functions, void ratio in relation to stress (alternatively pore pressure in relation to stress), and void ratio in relation to hydraulic conductivity; (c) discharge history; and (d) hydraulic boundary conditions. Of great use are survey data of tailings surface settlement after placement of the interim cover and data on settlement response from field tests. In this respect, it must be pointed out that the largest settlements do not necessarily occur at the deepest point of the slimes zone if heterogeneous sand-clay tailings layers are present.

3.2 TAILINGS BEHAVIOR FUNCTIONS

For material characterization, the results of the oedometer testing on borehole samples are needed to derive the compression index, C_c. On the basis of data for Culmitzsch A, the following relationship between the void ratio, e, and effective stress, σ', applies:

$$e = -C_c\,(\log \sigma') + e_0 \qquad (1)$$

where C_c and e_0 are the compression index and measured void ratio, respectively. For the empirical relationship of the void ratio, e, and hydraulic conductivity, k, a logarithmic function, is also used [7]:

$$e = C_k\,(\log \mathrm{k}) + e(k_0) \qquad (2)$$

The best validated data of the derived material parameters, C_e, e_0, C_k , and $e(k_0)$ come from Culmitzsch (Table 3) [8].

TABLE 3. Ranges of material parameters C_c , e_0, and $k(e_0)$ in the WISMUT impoundments

	Compres. Index, C_c [–]	Void ratio, e_0 [–]	Hydr.conduct., $e(k_o)$ [m/s]
Culmitzsch A			
Slimes	0.60 to 0.80	> 3.0	2 to 6 x 10^{-8}
Transition zone	0.40 to 0.60	1.7 to 3.0	1 to 2 x 10^{-8}
Sandy beach	< 0.10	0.5 to 1.1	10^{-4}
Trünzig B			
Slimes	(0.56)	(3.01)	(1x10-9 to 5x10-8)
Helmsdorf			
Slimes	0.50	1.5 to 3.0	1x10-9 to 2x10-8
Transition zone	0.20	0.7 to 2.0	1x10-9to 5x10-7
Sandy beach	0.05	0.55 to 1.0	1x10-4 to 5x10-7

The curves in Figure 3 illustrate the strong dependence of the slimes void ratio on the effective stress in contrast to the sandy tailings where the void ratio is insensitive to an increase of the effective stress over several orders of magnitude. The hydraulic conductivity of tailings can vary by four orders of magnitude for the common range of void ratios of 0.5 to 3.5.

The increase in load on the tailings surface first results in an equivalent increase of pore pressure over the previously static level. With dissipation of this excess pore pressure, consolidation sets in, and the additional stress that is due to the increased load is gradually transferred to the soil skeleton, thus effecting settlement. Owing to the low hydraulic conductivity of the fine tailings, the rate of pore pressure dissipation and, thus, of settlement is slow in the slimes zone.

3.3 DISCHARGE/DEPOSITION HISTORY MATCHING OF MODEL RESULTS

The reconstruction of time-dependent consolidation during discharge and standstill periods of the impoundment is needed for a realistic selection of the initial conditions to solve the nonlinear equation that describes the settlement. By history matching, the consolidation model is calibrated at the scale of the whole tailings impoundment. To predict future settlement under the load of the remediation cover, the actual void ratio (and/or pore pressure) profiles have to be known as well.

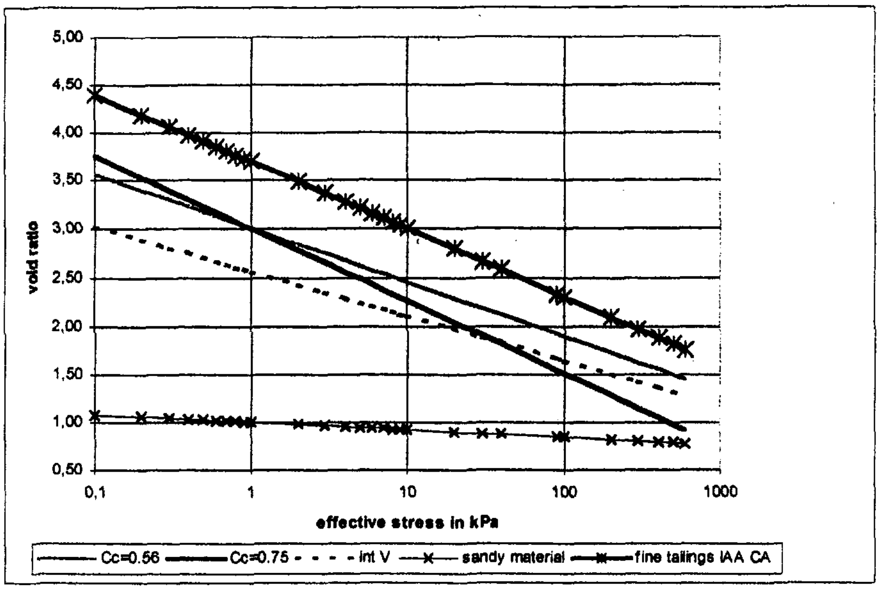

Figure 3. The void ratio expressed in relation to effective stress relationship for various tailings materials. The curve with $C_c = 0.56$ is based on the values from the Trünzig impoundment. The curve with C_c=0.75 was calculated to describe and demonstrate the sensitivity of the results from the compression index parameter. The rest of the curves illustrate the geomechanical behavior of the tailings at Culmitzsch A. The curve marked as "int V" corresponds to the tailings material in the transition zone in IAA Culmitzsch A [8].

3.4 SETTLEMENT MODELLING

The settlement modelling performed for Helmsdorf and Culmitzsch A showed that the classical linear soil-settlement theory (Terzaghi) developed for sandy material cannot adequately describe the geomechanical behavior of the fine tailings. For this reason, calculations of tailings settlement have to be done by nonlinear, finite strain (NLFS) models, such as ACCUMV and FSCONSOL, or the advanced NLFS codes developed at WISMUT.

The NLFS codes can account for the time-dependent effect of large strains on soft materials and provide the required accuracy. The changes in material properties induced by the large strain are taken into consideration by updating material properties with each time step. Furthermore, the WISMUT NLFS code can account for the radial effect of vertical drains used for pore pressure dissipation.

The simulation is performed for a particular place or profile of the impoundment. The observed drainage (or lack of drainage) at the base of the tailings deposit defines the lower boundary conditions that determine whether excess pore pressure can develop or not. The simulated pore pressure profiles at Helmsdorf (Figure 2) at the end of the filling and settlement period indicate that the fine slimes had an excess pore pressure of up to ~40 kPa (at the base of HW11) because of the low hydraulic conductivity of the tailings. The excess pore pressure was able to dissipate at the base of HW 12 because of the permeable subsurface conditions.

The results of the simulation are the time-dependent change in the thickness of the tailings deposit, change in the average void ratio with time at a specific location, or change in the void ratio with depth for a specific date. On the basis of the progress of consolidation, filling and grading of the interim cover surface can be scheduled.

The timing of the placement of the final cover over soft slimes must consider that (a) the compaction of the low-permeability cover with heavy equipment must happen at a stage of consolidation when the increase of shear strength of the underlying slimes is stabilized to a depth capable of accommodating the deformations induced by compaction without cracking; (b) if the low-permeability layer is placed while the slimes are still consolidating, the low-permeability layer must be sufficiently flexible to tolerate the differential settlements without cracking; (c) construction must not damage the functionality of the drainage material placed below the low-permeability layer; (d) the ultimate drainage pattern must ensure that no ponding on the low-permeability layer takes place; and (e) the long-term settlement of the low-permeability layer and any drainage layers must not alter the slope of the drainage pattern because of consolidation of slimes.

Because of the slow consolidation of the slimes, enhancement measures will be required for a timely remediation in order to be able to begin construction of the final cover to be placed on a consolidated tailings deposit (i.e., after approximately 90% of settlement is achieved).

4. Evaluation of the Remediation Strategies

Evaluation of the potential approaches to remediation involved the following individual aspects:

- *Hydrologic assessment.* Because of the potential hydrological consequences of a dam failure and proximity of the Helmsdorf and Dänkritz impoundments to population centers (approximately 500 m) and public roads (approximately 100 m), the dry cover remediation option was confirmed as the optimal long-term solution.
- *Water balance.* Exact knowledge of water input and output from the tailings impoundments was required for evaluation of water treatment needs. The seepage flow due to tailings consolidation had to be added to the standard water balance components.
- *Embankment slope stability.* In the case of Helmsdorf, hydrogeological modelling showed that the phreatic surface in the tailings can be reduced to a level that provides sufficient safety for the main dam in the present state and during flattening of the embankment.
- *Seismic stability.* The seismic stability of the Helmsdorf main dam was investigated and certified stable under both static and dynamic load conditions.

- *Drainage system.* After decommissioning of the tailings facility, the functioning of the drains was rechecked and measures were taken to keep them unclogged. Automatic flow recorders were installed to monitor seepage.
- *Erosion protection.* The most important erosion protection measure is the flattening of the external slopes of the embankments. The final shape geometry is designed to achieve a desired drainage pattern. Otherwise, planting of suitable vegetation on the slopes and tailings surface provides erosion control.
- *Dewatering and interim cover placement.* While keeping in mind the rehabilitation of the facility as a whole, the evaluation had to specifically account for the different geomechanical behaviors of the sandy beach, slimes, and intermediate zones. The purpose of the interim cover is to provide a radiological protection layer, induce tailings consolidation, create a trafficable surface, and provide the foundation for the final cover.
- *Final cover.* Current designs propose a multiple-layer, multiple-function configuration for the final cover system made of mineral soils [9]. A low-permeability layer consisting of clay material reduces infiltration into the tailings; the cover layers are designed to protect the cover as a whole.
- *Landscaping.* Landscaping is carried out with the goal of fitting the impoundment into the natural landform, which will enhance the development of a natural succession of indigenous plants.

A probabilistic evaluation of various combinations of remediation technologies and the duration of individual steps along with costs was conducted to evaluate the sequencing of remedial measures [10]. The study showed that the remediation can be speeded up considerably by use of deep drains in the slimes zone or, alternatively, that using less complex approaches over a longer period of remediation can save substantial costs.

The time-dependent evaluation of the complete sequence of steps needed for tailings impoundment remediation identified the slow rate of fine tailings settlement to be on the critical path, so that it could affect the overall duration of the remediation project. The reason for this significant effect is that the integrity of the final cover requires a well-consolidated base to prevent ponding and inhibit infiltration. The development of damage to the final cover by differential settlement would mean noncompliance with the objectives of the rehabilitation and may lead to increased seepage treatment costs.

5. Implementation of Remedial Measures and Technologies Used

When the pond water decreases, the predominantly sandy tailings close to the embankment consolidate quickly enough (within weeks) to provide beaches with sufficient strength to carry the load of the machinery ($>10\ kN/m^2$) needed to place the interim cover. To operate machinery on the tailings surface, a safety factor of $\eta \geq 1.3$ is required. For the construction of the interim cover, material from nearby waste rock dumps is used.

The increased risk of failure during construction of the dry cover on the low shear-strength tailings of the transition zone was handled successfully by adjustment of the rate of advancement of the cover (i.e., by decreasing the loading rates, using geotextile, polypropylene geogrid, and 4-to-5 or 8-m-deep vertical drains to accelerate pore pressure dissipation).

For installation of the shallow drains in the interim zone, a small lightweight pushing frame is used that moves on rails supported by boards placed on geogrid/geotextile on the soft slimes. A 1-m-cover layer is then advanced using a medium-sized dozer. This system has been effective when in situ total shear strengths are greater than 5 kPa. The cover advancement rates achieved on the tailings beaches and transition zones of Trünzig A and Helmsdorf were 0.5 to 2 m/day, depending on the shear strength of the tailings surface.

The technology described was tested at Helmsdorf during 1991/1993 and at the Trünzig A basin during 1994/1995. The placement of the interim cover on the Trünzig A tailings pond was successfully completed in 1995. For more extensive areas of deep slimes and steep transition zones at Helmsdorf, this technology is not suitable [11].

To avoid deeper-seated pond-slope failures under steep transition zones, the use of drain mattresses is envisaged. Installation of intermediate or deep drains may be required where high ultimate cover loads will be placed.

Advancing over slimes of lower shear strengths (< 5 kPa) will require the use of thinner layers, lighter equipment, or other methods currently under development.

A field trial of placing waste rock material subaqueously onto slimes to improve trafficability for the purpose of placing covers is currently under way at Helmsdorf. Above that, another layer is placed subaqueously on a geotextile layer. Initially, only the buoyant weight of the sand will contribute to the load on the slimes. Once the pond level can be decreased, the total weight of the emplaced rock becomes effective. The advantage of this rather complex approach is that the total weight of the initial layer can be applied to the slimes in two increments with no construction equipment load.

6. Control of Remediation Performance

6.1 EMISSIONS AND DUST

Control of emissions and dust was the most urgent task of the investigation and assessment of the tailings ponds during 1990 to 1993. Immediate remedial measures were necessary because the dose equivalent in the vicinity of tailings was estimated to reach 5 mSv/a above the background level (admissible level is 1 mSv/a). The high exposure rate was due to radon emissions and suspension of contaminated dust by the wind. The radiological health risks were successfully contained by immediate placement of an interim cover. This remedial intervention resolved the problem of dusting, decreased the radon emission from approximately 3 Bq/m^2s to less than 0.5 Bq/m^2s, and reduced external exposure from approximately 4 µSv/h to 0.4 µSv/h. As the pond water decreases, the interim cover is placed on the exposed areas, thus controlling radon emission or dusting.

6.2 SEEPAGE AND DISCHARGE WATER QUALITY

The initial investigations also revealed that contaminated seepage was migrating from the tailings ponds into the ground water and receiving streams. As an immediate control measure, the seepage collection facilities were reconstructed and upgraded. The upgraded seepage collection system at the toe of the Helmsdorf main dam now catches approximately 90% (2×10^6 m^3/year) of the contaminated seepage. For treatment of the tailings pond and seepage water, a water treatment plant was designed and commissioned at Helmsdorf in 1995. Passive water treatment methods are tested for long-term treatment of the seepage from the tailings impoundment.

6.3 SURVEILLANCE AND MONITORING

Immediately upon decommissioning of the tailings facilities, the sites were fenced to control unauthorized access. The undrained shear strength, excess pore pressure, and consolidation are monitored during cover placement for safety purposes. Long-term environmental performance (i.e., the atmospheric and aquatic impacts, as well as the geochemical conditions of the rehabilitated tailing ponds) will be monitored. Long-term surveillance and monitoring plans are being developed.

7. Conclusions

The sequence of tailings remediation steps followed at WISMUT to achieve the overall rehabilitation goals comprises (1) removal of pond water; (2) placement of an interim cover; (3) tailings consolidation enhancement by dewatering; (4) flattening of embankments and dams; (5) filling, grading, and shaping of the tailings surface; and (6) construction of the final cover.

Proven technologies for remediation of the tailings beaches and intermediate tailings zones are available at WISMUT. The present focus is on the development and testing of the technologies needed for remediation of the soft tailings.

Because of the slow consolidation of fine tailings, the approach to their remediation has a substantial effect on the costs and duration of a remediation project. In selecting the tailings remediation strategy, therefore, it is essential to consider the cost/time trade-off of the combination, sequencing and timing of remedial steps, and enhancement measures.

The slow consolidation of fine tailings and the positive water balance of the tailings impoundments make the use of tailings dewatering measures necessary. However, dewatering of the fine tailings by deep drains would unavoidably lead to increased construction costs; while, on the other hand, slow consolidation of the unstabilized fine tailings would result in long-lasting release of contaminated seepage from the impoundment, thus causing higher water treatment costs in the long run. Because a walk-away rehabilitation of tailings impoundments would be prohibitively expensive, it has to be taken into consideration that, after remediation, an object of restricted use will be left behind. This will require seepage water treatment, residue management, monitoring, and maintenance. For these reasons, it is necessary to raise the question of care for the remediated tailings impoundments at an early stage of the remediation project to minimize subsequent maintenance costs.

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POST-REMEDIATION MONITORING AND MAINTENANCE EXPERIENCE AT URANIUM MILL TAILINGS SITES, UNITED STATES

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1. Introduction

There are more than 40 uranium mill tailings disposal sites in the continental United States. Remediation has been completed at 23 of these sites. The remaining sites will be remediated by the year 2020 or thereafter. Remedial action at these sites includes decontamination of the mill and surrounding land, and isolation of tailings and associated contaminated materials in specially designed disposal cells. The oldest disposal cells were completed in 1985–1986 and are now 12 to 13 years old. This paper provides a summary of U.S. Department of Energy (DOE) experience at several of these disposal cells.

Surveillance, monitoring, and maintenance of completed uranium mill tailings disposal cells in the United States is the responsibility of the DOE Long-Term Surveillance and Monitoring (LTSM) Program, established in Colorado at the DOE Grand Junction Office (GJO) in 1988.

The mission of the LTSM Program is (1) to assume long-term responsibility for the disposal sites after they are remediated and (2) to ensure regulatory compliance and long-term integrity and safety, or long-term performance, of the disposal cells; or, in other words, to ensure the long-term isolation of tailings to protect public health and the environment. This responsibility includes annual site inspections, site security, ground-water monitoring, maintenance and repair, emergency response, annual reports, records management, and communication with regulators, local governments, and the public. At selected sites, radon monitoring and operation of water treatment and leachate collection systems will also be required.

2. Sites

In most cases, disposal cells are at or near former uranium mills built near the uranium mines. In a few cases, such as sites in Pennsylvania, ore was transported a long distance to make use of an existing milling facility. Most sites are in relatively remote, rural areas where few people live. Some sites are notably urban.

Disposal cells vary in size from 4 hectares (ha) to more than 300 ha. Volumes of tailings in the disposal cells range from approximately 70,000 cubic meters (m^3) to 19,800,000 m^3. Levels of radioactivity in the disposal cells range from approximately 40 curies (Ci) to 128,000 Ci (estimated total activity).

Contaminants in the disposal cells are low-level radioactive wastes (e.g., tailings, slimes from settling ponds, contaminated equipment, building materials, and soils). Primary contaminants are uranium and thorium daughters, nitrate, and heavy metals that are considered toxic. A few sites also contain small quantities of polychlorinated byphenyls (PCBs) and asbestos. (Chemicals, such as PCBs and asbestos, are placed in uranium mill tailings disposal cells only if they are associated with the tailings as "process-related wastes.")

Most disposal cells are surface impoundments, hill-like, with positive relief. Natural swales or small valleys were used for some sites, in which case the disposal cell may be low profile with little relief.

No uranium mill tailings disposal sites in the United States are in geographic settings exactly like Sillamäe. Most are in the semiarid western United States. None are along a coastline or in areas with a maritime climate. Some are, however, in humid climates or climates that are seasonably humid. In this paper, the focus is on disposal cells in areas where temperatures and precipitation are most like the climate in Sillamäe.

3. Regulatory Environment

Uranium mill tailings disposal cells in the United States, remediated under the Uranium Mill Tailings Radiation Control Act (UMTRCA) and regulated by the U.S. Nuclear Regulatory Commission (NRC), are under four principal regulatory constraints: (1) longevity, (2) passive design, (3) limitations on escape of radon, and (4) limitations on the leaching of radioactive and metal contaminants.

3.1 LONGEVITY

Disposal cells are to last for 1,000 years; if that is not achievable, for a minimum of 200 years. The 200-to-1,000-year design life is a political objective, as well as a technical objective. It is a political objective in that no government has lasted for 1,000 years without change or periods of instability. Whether or not any government, or succession of governments, can provide "institutional control" for 200 to 1,000 years remains to be seen.

3.2 PASSIVE, NO-MAINTENANCE DESIGN

The second constraint is a *passive* design, a design for which there is reasonable expectation that the disposal cell will survive for 200 to 1,000 years *without the need for active or planned maintenance*. What constitutes a passive, no-maintenance design is open to interpretation; the concept has been applied somewhat differently at some of the sites. The LTSM experience is that some maintenance is required very soon after construction at most sites. None of this work was anticipated. Accordingly, the cost was also not anticipated. These costs are likely to continue and may increase, because many, if not most, disposal cells may not survive in their intended condition for 200 years without this maintenance and repair.

Disposal cells are conceptually closed systems. They are built to ***contain*** waste materials indefinitely. In reality, natural processes that tend to "open" the system quickly develop. The entropy argument is that it takes energy to build such an ordered system; the system will begin to deteriorate and return to disorder once the energy is withdrawn. The preferred strategy is to design and build disposal cells that take advantage of inexorable long-term natural changes. To ensure the longevity of the disposal cell and to control the cost or maintenance and repair, disposal cells should be designed to take advantage of natural changes rather than be a barrier against them.

Elected and administrative officials at all levels of government, many of whom are not technically trained, may be disappointed to learn that the cost of a disposal cell is not just the one-time cost of construction. Funding to maintain the site must also be anticipated and provided. These costs may be hard to estimate or forecast accurately because of the many variables involved, at least some of which may not been foreseen or anticipated in the engineering design.

Lesson Learned Number 1

Maintenance and repair may be required in the years after the disposal cell is built. This possibility must be recognized and funding provided, if, under the best of circumstances, the disposal cell is to survive, as built, for more than a few years.

3.3 RADON RELEASE

The regulatory limit on release of radon to the atmosphere is 20 picocuries per square meter per second (pCi $m^{-2}s^{-1}$). This measurement can be area averaged over the entire surface of the disposal cell. An occasional higher reading or "hot spot" is allowed. To meet this standard, the disposal cell must have a cover over the tailings that limits the release of radon to the 20 pCi $m^{-2}s^{-1}$ standard for the lifetime of the disposal cell. Measurements of radon flux immediately after disposal cells are built are usually less than 1 pCi $m^{-2}s^{-1}$.

3.4 RADIOACTIVE MATERIALS AND METALS RELEASE

In addition to limiting the release of radon, the design of the disposal cell must achieve long-term containment of (1) radioactive materials, chiefly uranium daughters, and (2) associated toxic metals. If meteoric water flows through the disposal cell cover, leaching will doubtless occur; and contaminants, as leachates, will move into surrounding soils, ground water, and surface waters. (Contaminants can also be released by (1) uptake and transport through the vascular system of deep-rooted plants and (2) tailings brought to the surface by burrowing animals. Neither is a problem at U.S. sites at this time.)

Standards, referred to as Maximum Concentration Limits (MCLs) for release of radioactive and toxic metals, are established by the U.S. Environmental Protection Agency (EPA). Ground-water monitoring for contaminants with MCLs is required at some sites for lengths of time that vary from a few years to indefinitely. This too is a cost item that must be anticipated and funded.

Increasing concentrations of contaminants, concentrations above MCLs, are occurring in ground water at two disposal sites. The problem is under study. At this time, the problem is not related to maintenance or repair of the cover materials that overlie the tailings.

4. Strategy

The strategy to achieve the four constraints discussed previously is to preserve the radon barrier that is part of the engineered cover that overlies the tailings. The cover, including the radon barrier, must survive erosion, prevent release of radon, and prevent meteoric water from flowing through the disposal cell and leaching contaminants to surrounding ground water and soil for the required 200 to 1,000 years. (At some sites, the radon barrier is referred to as a "compacted soil layer" [CSL]).

5. Disposal Cell Design

The surface of the disposal cell may be (1) entirely rock covered (in dry climates), (2) vegetated and partially rock covered, or (3) completely vegetated (in humid climates) to prevent erosion. Riprap is used at many sites to prevent erosion of underlying layers, including the radon barrier. The size of the rock, or $D_{50,}$ is determined by the sheet flow that would result from the Probable Maximum Precipitation (PMP) or Probable Maximum Flood (PMF) event. D_{50} is the diameter of rock, such that half, or 50 percent, of the rock, by weight, must be larger. Catastrophic floods or precipitation worse than the design PMP or PMF is considered an acceptably rare risk at U.S. sites.

In humid climates, plants growing over a thick soil layer are considered a reliable means of erosion protection for the 200-to-1,000-year lifetime of the disposal cell. Deep-penetrating root systems of some plants continue to be a concern and the design must take account of them. Changes in climate, due perhaps to global warming, might alter the vegetation; but the change is expected to occur slowly. Geographic shifts in vegetation may actually lag climate change by decades or even centuries. Under most scenarios, the new plant succession would likely be equally successful in preventing erosion, unless the climate change were in the direction of excessive drying.

Below the erosion protection layer, there may be several underlying layers above the radon barrier; each layer has a specific purpose. The more complex designs may incorporate such layers as lateral drainage, bedding, root protection, frost protection, and capillary barrier layers above the all-important radon barrier. These layers, together with the radon barrier, constitute the "cover" that overlies the buried tailings.

The radon barrier has two important functions: (1) to limit the release of radon into the atmosphere below the 20-pCi $m^{-2}s^{-1}$ standard and (2) to prevent meteoric water from entering the disposal cell and leaching contaminants to the surrounding environment (e.g., ground water, surface water, and soils).

The radon barrier is, typically, a clay-rich, naturally occurring soil that can be compacted to a saturated hydraulic conductivity value, K_{sat}, of 1×10^{-7} cm/s or less. The 1×10^{-7} cm/s value is a U.S. EPA standard. This value is empirical and based on the compaction that can generally be achieved in naturally occurring clay-rich soils. Bentonite is mixed with the clay-rich soil at some sites where sufficient quantity of compactible soil is not locally available or to reduce the cost of construction by reducing the thickness of the radon barrier. In semiarid regions, the radon barrier is typically "dry" or unsaturated. In humid regions, the radon barrier is more likely to be saturated, at least seasonally. At some sites, evapotranspiration may be sufficient to dry out the radon barrier, in whole or in part, at different times during the year. Saturated, seasonally saturated, or unsaturated conditions are acceptable as long as the flow of meteoric water through the radon barrier, when it occurs, is at rates of 1×10^{-7} cm/s or less. Should that rate increase over time, it would be a concern.

Thickness of the radon barrier varies from disposal cell to disposal cell and ranges from about 45 cm to 215 cm. The thicker radon barriers were installed at the earlier and now oldest disposal cells. Such thickness was subsequently determined to be excessively conservative. Thinner, 45-cm-thick radon barriers are typical of the newer sites. (Required thickness of the radon barrier is computed by using the NRC RADON model. An earlier version of this model, called RAECON, was used to determine the thickness of the radon barrier at the earlier sites. This model has been refined several times. Refinements have allowed thinner radon barriers to be installed in the newer covers.)

Protection of the radon barrier from erosion and root damage is the primary challenge to long-term performance and regulatory compliance of the disposal cells.

6. Biointrusion

Biointrusion includes animal burrowing and encroachment of deep-rooted plants on the disposal cell. So far, burrowing has been only a potential or hypothetical problem. It has occurred at a few sites, but the burrowing was not extensive and the animals did not establish permanent colonies on the disposal cell.

Biointrusion by deep-rooted plants is a serious concern at several disposal cells and may eventually be a concern at most. If roots perforate the radon barrier in sufficient number, the design K_{sat}, 1×10^{-7} cm/s, is likely to increase, and the initial relative impermeability achieved by compaction will be compromised.

The Burrell, Pennsylvania, disposal cell was completed in 1987. Field measurements at the Burrell disposal cell show that plant roots (and perhaps some related soil-forming factors) have increased the K_{sat} to 1×10^{-5} cm/s. Comparative measurements taken at a nearby analog site, a historic site undisturbed since the late 1800s, show that the plant succession (hardwood forest with little understory) is likely to increase the K_{sat} to 1 x 10^{-4} cm/s in a period of 100 years or so.

Values of K_{sat} in the range of 1 x 10^{-5} to perhaps 1 x 10^{-4} indicate that the Burrell disposal cell is, technically, out of compliance with respect to the design specification of K_{sat} = 1 x 10^{-7} cm/s. This is a concern. The goal is to design, build, and maintain sites so that they perform to the design specification. What are the consequence, in terms of risk, if they do not?

At Burrell, attempts have been made to control vegetation so that the K_{sat} value does not degrade further. These attempts have not been successful. Control has relied on agricultural-strength systemic herbicides, which themselves are hazardous chemicals. The plants in the humid climate at Burrell are aggressive; the "kill," using herbicides, has always been incomplete and short-lived. The same plants often return within the same growing season.

Continued use of herbicides at this site is no longer acceptable. Not only are the herbicides ineffective, but the chemicals are also hazardous and constitute an environmental risk in themselves. Use of hazardous chemicals on such a scale is to introduce a new risk in an attempt to control an existing risk. Clearly, both risks need to be evaluated. (Nor is it certain that the application of these chemicals on an annual basis for a minimum of the next 188 years is institutionally sustainable.)

The proposed solution is risk assessment. Risk assessment, currently under way, is to determine if the measured and expected increases in the K_{sat} values actually pose a risk to public health and the environment. Authors of the risk assessment will examine (1) rates of water movement through the radon barrier as it degrades in response to root penetration and (2) the concentrations of radon and leachable contaminants that may leave the disposal cell as K_{sat} values increase to the anticipated 1 x 10^{-4} cm/s value. Pathways for these contaminants to reach the biosphere will be then identified and evaluated and exposures will be calculated.

It is expected that, in the worst case, the pathways-based risk will be acceptably low because the disposal cell does not contain high levels of radioactivity. If so, DOE will propose an end to vegetation control and acceptance of the disposal cell with increased permeability of the radon barrier. If NRC and the stakeholders agree that risk is acceptably low, the site will be allowed to return to hardwood forest.

If risk(s) are unacceptably high, or if allowing the forest to reclaim the site without further intervention is unacceptable to the public, use of hazardous chemicals in an attempt to slow the encroachment of deep-rooted plants will likely continue despite the unplanned cost and potential problems associated with release of hazardous chemicals into the environment. Modification of the design of the disposal cell to compensate for the forecast increase in K_{sat} values may also be considered. An option might be to install a thick soil layer over the existing cover. This layer would allow the forest succession to occur without further damage to the radon barrier.

Lesson Learned Number 2
The succession of natural vegetation, especially in humid climates, is bound to occur and is difficult to control. Nature will have its way. Engineers of disposal cell designs must anticipate that (1) vegetation will encroach on the disposal cell, (2) some of this vegetation will be deep-rooted, and (3) the best, or least maintenance, design will either isolate the encroachment from the radon barrier or incorporate the vegetation to some advantage, such as using evapotranspiration to de-water the radon barrier, so that the vegetation is an asset to the long-term performance of the disposal cell.

7. Erosion

A disposal cell, properly designed, constructed, and maintained, will shed most of the water that falls on it as rain or snow. This can be a lot of water. A 13-cm rain, common at some U.S. sites, falling on a 39-ha cell produces approximately 50,000,000 liters of runoff. If the cell sheds most of this water, as it should, the water moving off the cell into both as-built and natural drainage channels will be substantially more than if the moisture falling on the cell soaked into the cell at the same rate as in naturally occurring soils. The potential for erosion from this increased runoff is significant.

Erosion caused by runoff has not occurred ***on*** any disposal cell so far. (Minor wind erosion [deflation] has occurred at two sites, sites that incorporate soil or a soil-rock mixture as part of the cover design. This erosion is now stabilized by the formation of an armored, desert pavement-like surface, and measurable deflation is no longer occurring.)

Disposal cells are designed to shed rainwater and snow melt efficiently without erosion, and so far they are doing so. The erosion that has caused problems is erosion around the edge or apron of the disposal cell or on neighboring property. The problem has been that erosion around the disposal cell, by headward migration of rills and gullies, has threatened to move into the edge of the disposal cell. DOE has intervened at several sites to prevent erosion from damaging the cover and exposing once-buried tailings. Without this intervention, it is not clear that erosion would have stabilized or stopped before exposing tailings or, worse, transporting tailings downstream away from the site.

The Lowman, Idaho, disposal cell is in a mountainous region with abundant snowfall and often heavy runoff from rapid snowmelt. Short but intense summer thunderstorms are also common. The climate is savannah with large pines and thick, grassy understory. The disposal cell was completed in 1991.

The Lowman disposal cell is covered with large, angular granite riprap. The graded and disturbed areas around the disposal cell are steeply sloped. Soil on these slopes is a friable, weathered bedrock (grus) with little organic material and no developed soil horizons. After final grading, these slopes were planted with annual grasses to hold the soil against erosion until native species could establish. The effort was not successful. Annual grasses sprouted the first season, but not thereafter. Native vegetation did not establish. Gullies began to form within one year.

Erosion became an issue in 1996, when gullies, some as large as 3-m deep and 10-m long, were noted. Headcuts in these gullies were migrating up the slope and rills above these headcuts began to reach into the edge of the disposal cell. Intervention was essential.

In October 1998, the eroded areas were regraded to fill the gullies and headcuts. Drainage terraces were installed to convey water across the slope and off site. The entire area was replanted by hydroseeding over a hydromulch. Cost to repair this erosion, approximately US $70,000, far exceeds what it would have cost to terrace and more carefully revegetate at the time the site was under construction in 1991.

Conceptually, erosion peripheral to the disposal cell is an "edge" effect. Areas around the edge of the disposal cell begin to erode because they were not properly designed, graded, revegetated, or otherwise protected from erosion. Erosion around the edge of the disposal cell increases until it directly threatens the disposal cell. This condition has occurred at several sites; the Lowman site is the most serious example.

Lesson Learned Number 3

Personnel experienced with erosion control techniques and revegetation of disturbed areas must be part of design and remedial action teams. These people must have authority to participate in the approval and acceptance of final site designs and to oversee the site while it is under construction.

Lesson Learned Number 4

Slopes around and below the disposal cell must be understood to be part of a geomorphic continuum. A change at one place along the slope or the construction of a positive feature, such as a disposal cell on a previously stable slope, will cause slope adjustments (erosion) and some of these will eventually threaten the disposal cell.

8. Degradation of Riprap

The Lakeview, Oregon, disposal cell was built in 1989. It is also in a mountainous area. Winters are wet; summers are dry except for thunderstorms. The radon barrier cell is protected from erosion by a rock-and-soil mixture on top of the disposal cell and by riprap on the long, west-facing side slope of the disposal cell. Plant encroachment on the rock and soil mixture on top of the disposal cell is a potential problem currently under study, but the bigger problem is the condition of the riprap on the side slope.

The riprap is a weathered olivine basalt. The design called for a riprap layer 15 cm thick with a D_{50} of 2.7 inches. Inspectors at the quarry noticed that some of the rock was cracked and crumbling as it came from the quarry wall. Much better rock, a densely crystalline basalt, was available from a quarry 150 km away. To contain cost and maintain the construction schedule, both of which would have been adversely affected by a 150-km haul, the decision was made to use the weathered, poor-quality basalt but to double the thickness of the riprap layer and to use somewhat larger rock than the minimum required. Doubling brought the as-built thickness to approximately 30 cm.

The riprap was installed in 1987. In 1994 it was noticed that numerous stones were cracking and splitting or spalling as they lay in place on the surface of the disposal cell. The rock was deteriorating under surface weathering conditions. Petrographic examination of the rock showed that the rock was microfractured throughout, the glassy matrix and feldspar crystals were cloudy and altering to clay, and olivine was altering to iron oxides along edges and fractures. The rock was weathering so fast that a procedure to measure the rock and monitor its decrease in size was instituted in 1996.

The design at Lakeview called for a D_{50} so that 50 percent of the stones, by weight, must be larger than 7 cm. Riprap 7 cm to 10 cm in diameter was installed to meet, conservatively, the 7-cm requirement. Measurement of the rock in 1997 and 1998 showed the mean diameter of the riprap had decreased, due to weathering, to about 7 cm. Thus, the riprap was at the lower limit of the design requirement. (Doubling the thickness of the poor quality riprap was not a good solution. Part of the function of large-diameter rock is to provide "roughness" to dissipate the energy of overland flow. A double thickness of poor quality rock eventually becomes a double thickness of small rock, and this does not achieve the required weight or roughness.)

The size of the riprap is now borderline. Weathering of the riprap is expected to continue. When the riprap breaks down to the point that the D_{50} is less than the design requirement, intervention may be necessary. Options include (1) recalculation of the statistical PMP to confirm no errors were made; (2) argument that the statistical PMP is inappropriate for the location of the disposal cell; (3) pathways-based risk assessment, as discussed previously for the disposal cell at Burrell; and (4) redesign and reconstruction of the disposal cell cover. The latter might include the addition of more, better quality riprap or installation of deep soil and revegetation. The cost for this option could be in the US $1,000,000 to US $2,000,000 range. Such cost, to maintain and repair a disposal cell after it is built, is unprecedented.

Lesson Learned Number 5
Use of poor or marginal quality materials is not consistent with a low- or no-maintenance disposal cell design.

9. Conclusions

A passive, no-maintenance disposal cell design is a worthy objective. It is difficult to achieve.

The disposal cells built in the U.S. for uranium mill tailings are so far successful. The oldest cells, built more than 12 years ago, are performing well. No tailings, once buried, have been exposed, and no contamination of ground water has been recorded as a result of leaching from the disposal cells. However, there have been problems, chiefly problems related to maintenance and protection of radon barriers as installed. Lessons learned at U.S. uranium mill tailings disposal cells may be transferable to Sillamäe and similar sites.

- Disposal cells are conceptually closed systems. In reality, natural processes, which tend to "open" the system, quickly develop. Energy is spent to construct the containment system; containment begins to deteriorate once the energy is withdrawn. Therefore, maintenance and repair will be required to preserve the life of the disposal cell and its ability to contain waste materials. Governments and agencies that provide environmental oversight must understand that maintenance will be required and that there must be a source of funding for this work if the disposal cell is to survive, under the best circumstances, for more than a few years.

- The engineering design for the disposal cell must anticipate the encroachment of vegetation, especially in humid climates. The natural succession of vegetation will occur and will be difficult to control. Engineering designs must anticipate that (1) vegetation will appear on the disposal cell, (2) some of this vegetation will be deep-rooted, and (3) the best (least maintenance) design will use the vegetation in a way that will be an asset to the long-term performance of the disposal cell. The preferred strategy is to design and build disposal cells to take advantage of long-term natural changes and natural processes, such as evapotranspiration. This is the only practical way to ensure the longevity of the disposal cell and to control maintenance and repair costs.
- Teams that design and build disposal cell must include personnel familiar with erosion control techniques, revegetation of disturbed areas, and evapotranspiration cover designs. These people must have authority to participate in the approval and acceptance of final site designs and to oversee the site while it is under construction.
- Slopes and drainage systems that surround disposal cells must be understood to be part of a geomorphic continuum. A change at one place along a previously stable stream bank or hillslope, such as the construction of an above-grade disposal cell with positive relief, is likely to cause or to accelerate erosion that may eventually threaten the disposal cell.
- Use of poor or marginal quality materials in disposal cell covers, such as poor quality rock for erosion control, is not consistent with a low- or no-maintenance disposal cell design.

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CURRENT STATE OF REMEDIATION OF TAILINGS PONDS AND THEIR SURROUNDINGS IN HUNGARY

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1. General Description of Uranium Mining Activity

Exploration for uranium ore started in 1953 in Hungary. Soon after the first gamma radiation survey was conducted, it became clear that a significant uranium deposit existed near the town of Pécs. After intensive exploration work in 1954, a company was established (with the Hungarian name *Bauxitbánya Vállalat*) in 1955 for the development of uranium mining. In 1956, the name of the Company was changed to *Pécsi Uránbánya Vállalat*; and after starting mill operations in 1964, the Company name was changed to *Mecseki Ércbányászati Vállalat (Hungarian abbreviation MÉV,* Mecsek Ore Mining Company).

In 1990-1992, the Company was reorganised into smaller limited companies. One of them, MECSEKURAN Ltd., became responsible for the mining and processing of uranium ore. Because of high production costs, the government decided to shut down the mines and the mill at the end of 1997; and processing of uranium ore was ended in 1997. The former company is responsible for the remediation work, which is why the new name of the company is Mecsek Ore Environmental Protection Joint-stock Company (in Hungarian *MECSEKÉRC Környezetvédelmi Rt.*).

The total quantity of uranium exported from this region is about 1.1 percent of total world production.

2. General Process Flow for Treatment of Mined Rock

The mined out rock was radiometrically sorted into three groups:

- Commercial mill-grade ore (U > 300 g/t)
- Low-grade ore (U = 100-300 g/t)
- Waste rock (U < 100 g/t)

The mill-grade ore was further enriched in a radiometric sorting station. Concentrate from this operation was processed in the mill, and the rejects were either dumped to the waste rock pile or processed by heap leaching with the low-grade ore.

The conventional mill process used sulfuric acid for leaching and counter-current extraction for removal of dissolved uranium. The uranium was obtained in the form of calcium diuranate. The barren pulp was neutralised by lime to a pH of 7-7.5 Tailings were discharged into the tailings ponds.

Low-grade ore and some of the rejects from the radiometric concentration station were crushed to <30 mm and hauled to the appropriately isolated leaching area where the ore was treated with sodium carbonate solution. The dissolved uranium was removed by anion exchange resin and, in the form of eluate, was directed to the mill, where it was processed into yellow cake. Mine water was also treated and recovered uranium from was directed to the mill.

3. Mined Rock Balance

Historical data were collected to obtain a total mass balance of rock and radionuclides that originated from the mining activity in Hungary.

3.1. MINED ROCK BALANCE

Mined rock amounted to 46.8 million metric tons (t) containing 23,083 t of uranium. From this quantity, according to available data, approximately 1.44 million t of ore with 2.195 t of uranium were exported directly to the Soviet Union. The waste rock piles contain 19.303 million t rock with a calculated average uranium concentration of 54 g/t. A total of 7.213 million t of low-grade ore was treated by heap leaching, while 18.886 million t was processed by a conventional acid mill process. The total material balance for all mined rock is presented in Table 1. The data are given for mass and the radioactivity activity as well. As shown in Table 1, 2,808 t of uranium remained after closing of the mining operation, mostly in the tailings ponds.

3.2. TAILINGS

During the operating period, one small experimental and two commercial tailings ponds were built for the tailings (TP I and TP II): one of them is a ring dike type (TP I), and the other is a valley type. The tailings ponds were built without isolation near the aquifer supplying the town of Pécs with drinking water. The area of TP I is 104 ha, while that of TP II is 56 ha. Table 2 summarizes the tailings pond data.

During the milling process, almost 2 million t of sulfuric acid, 623 thousand t of pyrolusite, and 83 thousand t of hydrochloric acid were consumed.

A total balance of the radioactive decay products shows that most of those mined are now in the tailings ponds and amounted to 2.38 x 10^{15} Bq from the U-238 chain and 9.89 x x 10^{13} Bq from the U-235 chain. This means that the following quantities of isotopes and others are in the tailings ponds:

TABLE 1. Material balance for mined rock

Item	Exported Ore	Processed in the Mill	Heap Leached	Waste Rock	Total
Mass of rocks, t	1,440,600	18,886,477	7,213,000	19,303,515	46,843,592
Avg. U-conc. in the rock, g/t	1,524	1001.57	130.7	53.34	492.78
Uranium, kg	2,195,000	18,916,221	942,776	1,029,715	23,083,712
Tailings*, t	0	20,397,300	7,213,000	-	27,610,300
U conc. in the wastes, g/t	-	67.88	~55	53.34	
Quantity of U in the wastes, kg		1,384,534	397,182	1,029,715	2,811,431
Utilised waste**					
Quantity of rocks, t		0	46,300	0	46,300
Uranium, kg			3,241		3,241
Decay products†, kg			6,570		6,570
Tailings deposited on the site					
Uranium, kg		1,384,534	393,941	1,029,715	2,808,190††
Decay products†, kg		18,916,221	936,206	1,029,715	20,882,142
Cross activity‡, Bq					
From U-238 chain		2.38×10^{15}	1.34×10^{14}	1.76×10^{14}	2.69×10^{15}
From U-235 chain		9.89×10^{13}	5.27×10^{12}	6.48×10^{12}	1.11×10^{14}
Total, Bq		2.48×10^{15}	1.39×10^{14}	1.83×10^{14}	2.80×10^{15}
Th-230, kg		327	16.2	17.8	361
Ra-226, kg		6.32	0.313	0.344	6.977
Pa-231, kg		6.05	0.3	0.33	6.68
Exported U, kg	2,195,000	17,534,987	545,594	-	20,298,500‡‡

* Processed ore

** Transferred from the sites

† Expressed in units of natural uranium equivalents

†† Uranium remaining on the sites

‡ Starting from uranium isotopes

‡‡ After deduction of uranium from heap leaching and including uranium from mine water treatment (22,919 kg)

- 1,384,534 kg natural uranium,
- 327 kg Th-230,
- 6.32 kg Ra-226, and
- 6.05 kg Pa-231.

The annual volume of processed ore and its distribution between the two tailings ponds is shown in Figure 1. The total quantity of tailings is 20.4 million t, of which 15.8 million t are found in TP I and 4.6 million t are in TP II.

TABLE 2. Quantity of tailings and radionuclides in tailings ponds

Item	**Milling**			**Total**
	Experimental Tailings Pond	**Tailings Pond No. 1**	**Tailings Pond No. 2**	
Mass of ore processed, t	~27,777	14,600,500	4,258,200	18,886,477
Avg. U-conc. in the processed ore, g/t	1,001.575	1,004.53	991.56	1,001.575
Uranium, kg	27,821	14,665,400	4,223,000	18,916,221
Tailings*, t	30,000	15,768,500	4,598,800	20,397,300
U conc. in the tailings, g/t	67.8	71	56	67.8
Quantity of U in the tailings, kg	2,034	1,125,400	257,100	1,384,534
Utilised tailings**				0
Tailings deposited on the site	30,000	15,768,500	4,598,800	20,397,300
Uranium, kg	2,034	1,125,400	257,100	1.384.534
Decay products†, kg				18,916,221
Cross activity‡, Bq				
From U-238 chain	1.85×10^{15}		5.29×10^{14}	2.38×10^{15}
From U-235 chain	7.69×10^{13}		2.2×10^{13}	9.89×10^{13}
Total, Bq	1.93×10^{13}		2.2×10^{13}	2.48×10^{13}
Th-230, kg				327
Ra-226, kg				6.32
Pa-231, kg				6.05
Produced U‡‡, kg				17,534,987

* Processed ore
** Transferred from the sites
† Expressed in units of natural uranium equivalents
†† Uranium remaining on the sites
‡ Starting from uranium isotopes
‡‡ In the form of yellow cake

3.3. SEEPAGE FROM TAILINGS PONDS

As the neutralisation of barren pulp was carried out at a pH of only 7.5, most of the magnesium dissolved during the leaching process remained in the process water and was also disposed of in the tailings ponds. As a consequence, the concentration of total dissolved solids (TDS) was high in this water. The TDS of process water and the quantity of the chemical compounds in this water are shown in Figures 2 and 3.

An average of more than 20 thousand t of TDS was discharged into the tailings ponds annually. The concentration of TDS in the process water was very high, reaching in some cases 40 g/l (e.g., in the first half of eighties). The water balance shows that approximately 20 million m^3 of water infiltrated from the tailings ponds into the surroundings. Table 3 presents the estimated composition of seepage from the tailings ponds. These data show that main pollutants were magnesium, sodium, sulfate, and

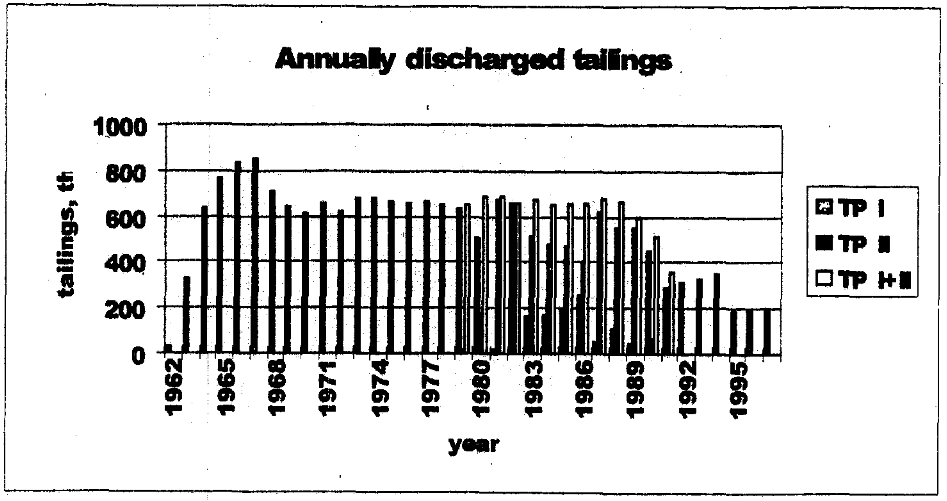

FIGURE 1. Distribution of tailings between the two tailings ponds

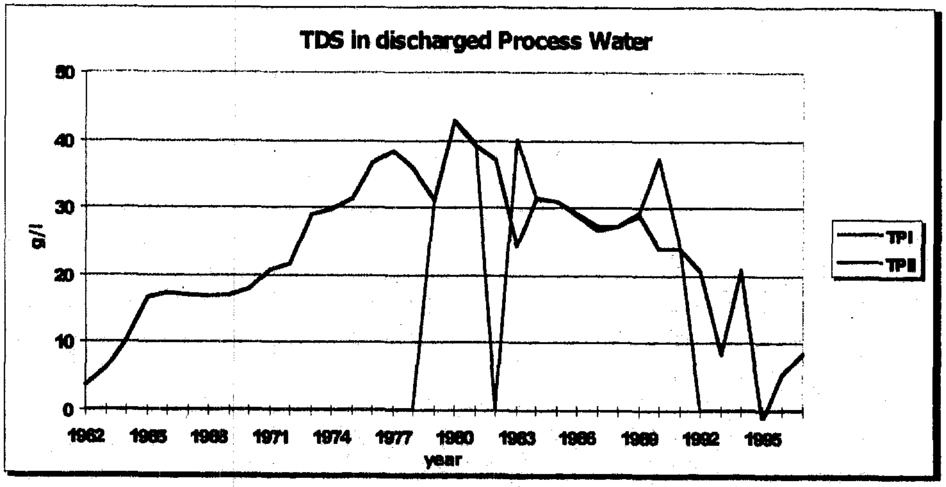

FIGURE 2. Concentration of TDS in process water

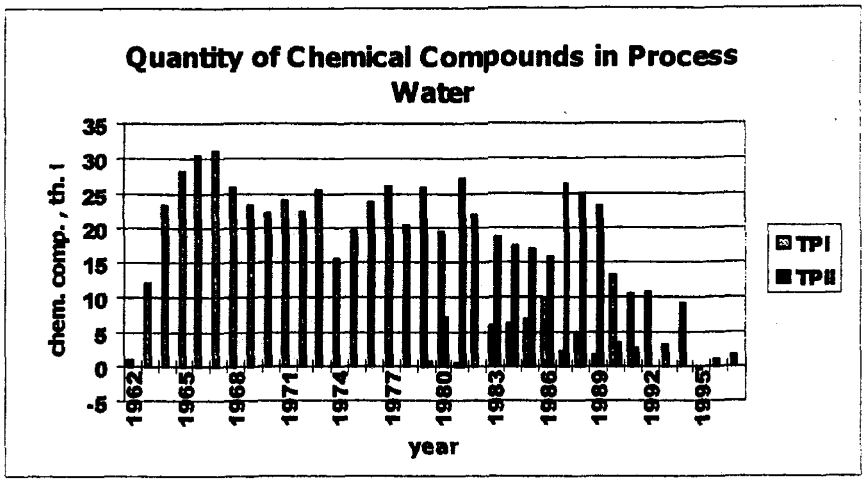

FIGURE 3. Annually discharged chemicals in process water

TABLE 3. Composition of seepage from tailings ponds

Na^+	K^+	Ca^{2+}	Mg^{2+}	Mg/Ca	Mn^{2+}	Cl^-	SO_4^{2-}	CO_3^{2-}	HCO_3^-	NO_3^-	TDS	pH	U	Ra
mg/l					mg/l								µg/l	Bq/l
1065	180	530	2800	5,5	710	2400	12400	<30	40	180	20400	7	44	520
meq/l														
46	5	27	239		13	68	258		0,7	3				

chloride ions. Heavy metal concentrations in the tailings water are very small and therefore are not indicated in the table.

3.4. POLLUTION OF GROUND WATER

From the data presented, it is clear that a very large amount of contaminants left the tailings ponds and infiltrated into the ground water. The presence of contaminants in shallow ground water and in a portion of the deeper ground water was detected by monitoring.

The amounts of TDS in shallow ground water are shown in Figure 4. It can be seen that the contaminants are dispersed over a huge area around the tailings ponds and the concentration of TDS reaches 18 g/l in some areas. The deeper second aquifer is much less polluted, but, nevertheless, it is evident that contaminants are moving toward it.

4. Ground water Restoration and Remediation of Tailings Ponds

Rehabilitation of tailings ponds usually means covering their surfaces to decrease radon release, gamma dose rate, and dust formation. In this case, it is also very important to carry out ground water restoration, because ground water in the vicinity of the tailings ponds is highly polluted with chemicals from the ponds. The aquifer from which the town of Pécs receives a significant portion of its drinking water is not protected by the hydrogeologic character of the site. The rehabilitation of the tailings ponds has to solve at least two problems:

- Restoration of ground water quality around the tailings ponds.
- Covering the tailings ponds to protect the environment from dispersion of radionuclides from their surfaces.

The overall remediation plan is now under development, but some work related to the general restoration of tailings ponds can be done now.

4.1. RESTORATION OF GROUND WATER

The tailings ponds were constructed without isolation. Water conductivity of the underlying rocks varies in the area around 10^{-6} to 10^{-7} cm/s. Therefore seepage from tailings ponds has taken place and the ground water has been polluted in the vicinity of these facilities. Water balance calculations show that an average of 1,600 m^3/d process water (with some water from precipitates) containing 32.56 t TDS leaked from the tailings ponds during the operating period of the mill. Ground water pollution in the vicinity of the tailings ponds is detected by monitoring wells. For the monitoring of ground water quality, there are many (more than 80) monitoring wells around the tailings ponds. The monitoring wells are constructed in pairs: one in the shallow water aquifer, the other one in the deeper water aquifer which has a direct connection to the drinking water supply for the area. Analyses of water samples show that the shallow water aquifer is highly polluted (TDS = 18-15 g/l) by tailings water. This can be seen in Figure 4. These data show that even the underlying deeper aquifer is polluted to some extent (TDS = 2-5 g/l). The geophysical mapping by vertical electrical sounding also indicates pollution with inorganic compounds at various depths. On the basis of analytical data from sampling of monitoring wells, the main pollutants are magnesium sulfate and sodium chloride. Uranium and radium are present only in small concentrations in the ground water. These data are generally in agreement with the composition of seepage presented in Table 3.

Because the contaminated ground water can reach the drinking water aquifer, it is extremely important to carry out ground water restoration work to protect the most important drinking water resources of the region. For water quality protection and restoration, possible technical measures have been considered (slurry wall, chemical active wall, water extracting system, etc.).

The ground water restoration work started recently. For the first step, extraction wells were constructed for shallow ground water. The water extracting pilot station consists of five wells constructed on the most contaminated site of the tailings ponds at

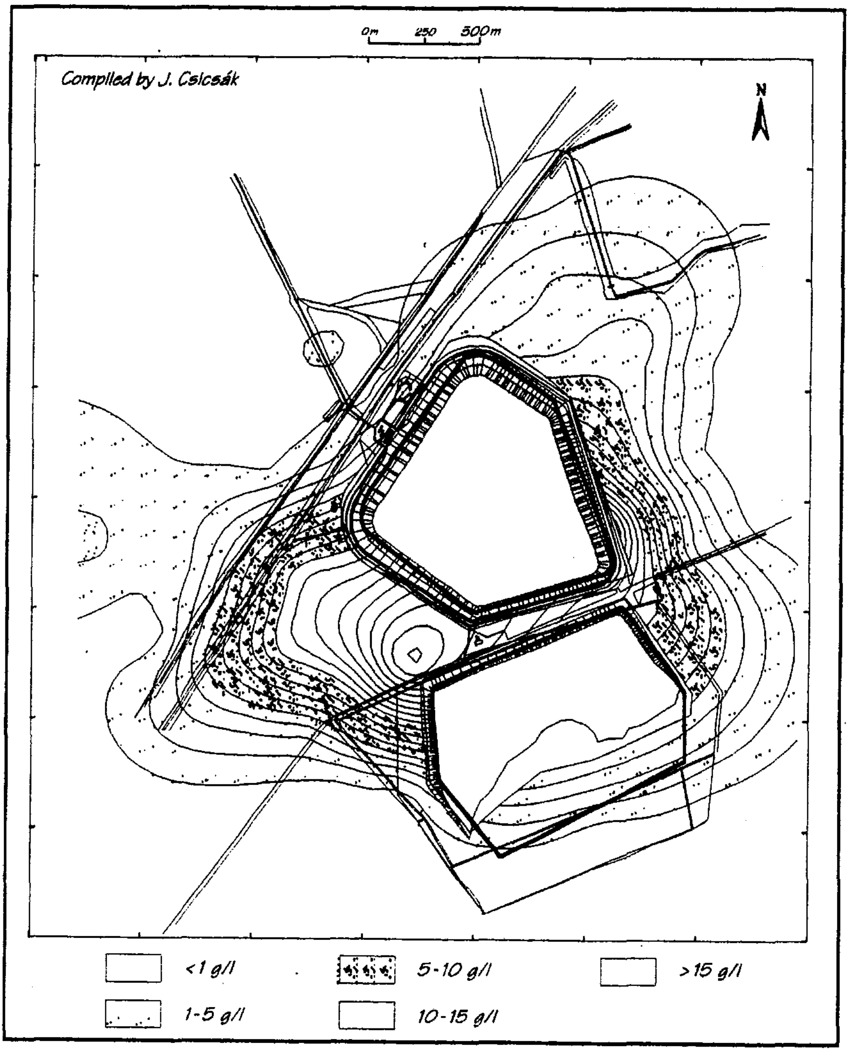

FIGURE 4. TDS content of shallow groundwater at the tailing ponds (g/l)
(first half-year of 1997)

points shown on Figure 5. Analytical data for extracted water from the first four months of operation of the pilot system is shown in Table 4. The extracted water contains ions that originated in the tailings process water. First of all, the magnesium (1.5 g/l), sodium (0.7 g/l), sulfate (6.6 g/l), and chloride (1.6 g/l) concentrations are high. It seems that the calcium concentration (0.6 g/l) is determined by gypsum solubility. Total TDS is about 13 g/l. It is important to point out that uranium and radium concentrations in extracted water are low enough that there is no danger of water contamination by these nuclides.

The experiment in general shows the possibility of removing the contaminated water, but more detailed lithological information is needed for construction of an effective large-scale water-extracting system. It is estimated that at least 1 million m^3/year of polluted water must be removed to protect the drinking water aquifer, but the real volume of water to be extracted annually for restoration of the site should be determined by appropriate modelling. The duration of water removal is not known at this time because of the lack of additional information (sorption capacity of soil for the contaminant in question, the natural dilution effect, etc.). Nevertheless, the ongoing test proved the possibility of ground water restoration by extraction of shallow ground water.

It is worth mentioning that some results from the sampling of monitoring wells and the ongoing water-removing experiment indicate that some self-restoration may be taking place. Potassium is very effectively sorbed by soil minerals, as its concentration in removed water decreased to <5 mg/l from 180 mg/l in tailings seepage. Radium sorption is also shown because its initial concentration in tailings water is >5 Bq/l but is only <0.1 Bq/l in extracted water. Uranium binding has also taken place to some extent.

From the point of view of general attenuation of the contaminant concentrations, it is important if the sorption of magnesium and sodium take place. In this respect, data indicate relatively high decreases of magnesium concentrations compared with that of calcium: while the magnesium concentration dropped from 2,900 mg/l (in seepage) to 1,569 mg/l (in extracted water), the calcium concentration remained the same (0.6 g/l) and the Mg/Ca ratio dropped from 5.5 to 2.95. These results can be explained by a cation-exchange process in soil:

$$\mathrm{Ca[soil]} + \mathrm{Mg\,SO_4} = \mathrm{CaSO_4} + \mathrm{Mg[soil]} \qquad (1)$$

where soil contains the anion group of minerals capable of ion exchange.

As the water becomes saturated with gypsum, most $CaSO_4$ obtained according to equation (1) precipitates from the solution, and the resulting effect is the decrease of the magnesium concentration and the TDS as well. Although this and possibly other physical chemical processes attenuate the contaminant concentrations around the tailings ponds, the restoration of ground water quality can be expected only if appropriate measures are undertaken.

Extracted water should be chemically treated to decrease the TDS before discharging it into the receiver. The principal process design of the water treatment technology is under development. The related investigations are sponsored by the International Atomic Energy Agency (IAEA).

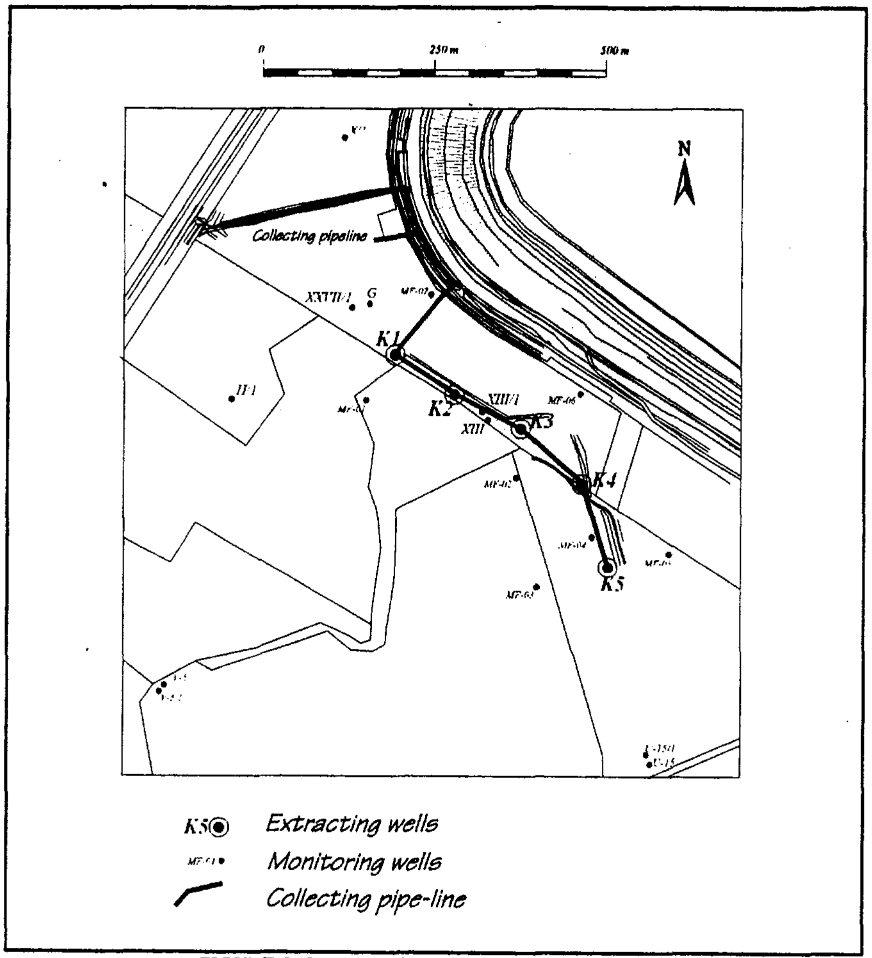

FIGURE 5. Location of water-extracting and monitoring wells

4.2. REMEDIATION OF TAILINGS PONDS

Before starting the remediation of tailings ponds, it is necessary to reach appropriate surface stabilisation and undertake methods for long-term stabilisation of the tailings if needed.

4.2.1. *Long-Term Stabilisation of Tailings Ponds*

The most important concern is the stabilisation of the slimy central part of TP I. The stability of the surface of the tailings pond following the removal of free water (the

TABLE 4. Chemistry of removed water

Date of sampling	Na^+	K^+	Ca^{2+}	Mg^{2+}	Mg/Ca	Cl^-	SO_4^{2-}	CO_3^{2-}	HCO_3^-	TDS	pH	U	Ra	Water pumped out
	mg/l					mg/l						µg/l	Bq/l	m³/day
21.05.98	803	<5	534	1673	3.13	1734	6980	<10	628	14400	7.1	20	0.094	
04.06.98	710	<5	600	1500	2.5	1700	6380	<10	694	12900	7.2	17	0.06	
30.06.98	720	<5	580	1550	2.67	1630	6890	<10	610	12300	7.1	12.5	0.053	
16.07.98	695	<5	520	1535	2.95	1595	6955	<10	622	13100	7.3	13	0.081	307.92
27.07.98	725	<5	514	1681	3.27	1648	6470	<10	631	13000	7.1	12.5	0.052	
31.07.98	725	<5	560	1475	2.63	1630	6130	<10	637	12600	7.0	12.5	0.038	
26.08.98	728	<5	536	1553	2,95	1613	6934	100	644	13200	6.9	12.0	0.074	
Average	729	<5	547.7	1566	2.95	1650	6688	<10	627	13067	7.13	16,66	0.06	
meq/l	31.70		27.55	129.14		46.54	138,92		10.27					

volume of which is now approximately 150,000 m³) will be insufficient for moving heavy equipment during covering. An appropriate technical method for stabilising the surface of this part of tailings ponds must be developed. The aim of the ongoing PHARE project is to solve this problem and design an optimal technical solution for the stabilisation of TP I. A stabilisation problem exists only on TP I; TP II has been out of operation some years and, therefore, it is almost dry.

Another question is the general dam stability. Dam stability will be addressed with the criteria for long-term stabilisation of tailings as part of the PHARE project, and the planning work can be started in 1999.

4.2.2. *Covering the Tailings Ponds*

An economic evaluation and risk analysis should be carried out to find the optimal solution to the question whether the smaller tailings pond (TP II) has to be relocated onto the bigger one or whether it is more reasonable to remediate them separately. This question is to be solved at the end of 1998 during the feasibility study.

The next question to be solved is the design of an appropriate structure to cover the tailings ponds. In support of the design, some field experiments were undertaken and some of them are still under way. The covering structure will be based on international recommendations and practice.

5. Conclusions

1. The rehabilitation of tailings ponds in Hungary is an important and urgent task as it can cause serious pollution of the main drinking water resource of the town of Pécs, population 180,000.

2. To protect this aquifer, it is necessary to perform direct ground water remediation by removing the polluted shallow ground water and perhaps incorporate other cleaning methods.
3. Ground water remediation must be performed in concert with the remediation of the tailings ponds, one of the important aspects of which is the design an optimal technical method for stabilisation of the tailings ponds. This task is to be solved as part of the PHARE project(s).
4. Data show that some self-restoration processes are taking place as the water originating in the tailings ponds moves through the soil.

THE ENVIRONMENTAL IMPACT OF FORMER URANIUM MINING ACTIVITIES AT THE NOVOVESKA HUTA SITE, SLOVAKIA

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Mining and processing activities around Novoveska Huta village are very old. A huge number of old mining dumps and other residues from mining and processing of copper ores are found in the area close to the village. Historical studies show that mining activities have taken place in this area from primeval times. In the second half of the nineteenth century, the copper ores had been completely exploited. From the beginning of the nineteenth century, gypsum and anhydrite were also mined. In the first half of the twentieth century, mining of sulfates and radioactive ores was developed.

The former state mining organisation, Jachymovske doly, and its successor organisations developed extensive mining and drilling exploration in the first half of this century. Their operations were focused on copper and uranium-molybdenum ores. More than 150 km of deep boreholes were drilled with an average depth of 200-300 m. The deepest borehole was 1960 m. During this time, about 90 km of underground mining shafts and adits were developed. The central part of the uranium deposit was explored to a depth of 650 m. During this period more than 760,000 tons of copper ores and 130,000 tons of uranium-molybdenum ores were exploited.

In 1990, the Czechoslovak Ministry for Fuels and Energy decided to end all research, exploration, and exploitation activities for copper and radioactive ores in Novoveska Huta. The mining facilities were to be decommissioned and remediated as necessary to minimise environmental liabilities. Most of the mining dumps were recultivated and the entries to adits were closed. Underground mining spaces under the outfall of Dubnica Brook were flooded. All low-grade ore dumps were evaluated by radiometric methods and the results documented. The report is in the archives of the former organisation Geofond in Bratislava.

Because the copper and uranium-molybdenum ores were processed outside the region (uranium ores were processed in Czech Republic and in the former Soviet Union, probably at the Sillamäe site) this locality was not impacted by processing activities. Before flooding of the underground mining spaces, all known springs, mining waters, surface waters and wells close to the former mining activities were analysed for pH, ^{222}Rn, ^{226}Ra, natural U, Cu, Ag, Hg, Pb, As, Se, V, Cd, Cr, Fe, Mn, ammonium, sulfates, nitrates, nitrites, dissolved solids, biological oxygen demand, and chemical consumption of oxygen by the manganometric method. In addition, the quantity of the waters was measured. Monitoring of these components continues and should continue for 10 years. Although after flooding the underground mining spaces some of the measured components increased (see Table 1), they are below the limits specified by Government order No 242/1993. The results of previously existing observations confirmed the stability of the output waters without significant changes in the quality of this waters. However, the quantity of some waters, e.g. output from the adit named *Vodna stolna* (Water adit) increased from 1 liter per second to 5 or 8 liters per second. From point of view of close connections of all underground spaces we can assume that this increase will continue in the future and will depend on seasonal rainfall.

In the last two years we had participated in the PHARE programme, "Remediation concepts for uranium mining operations in CEEC." It has been very helpful for review of environmental impacts and risks and for the effectiveness of recultivation after former mining activities. The experience at Novoveská Huta shows that it is possible to minimalize the environmental impact to an acceptable level.

TABLE 1. Underground waters in Novoveská Huta

Component (mg/liter)	**Recommended Value**	**Vodná Štôlna**		**Doloveika**		**Dubnicamost**		**Jánsky Potok**	
		During mining	**After flooding**	**During mining**	**After flooding**	**During mining**	**After flooding**	**During mining**	**After flooding**
BSK_5*	8	1.000	0.500	<0.20	<0.20	<0.20	0.900	1.800	0.200
CHSK Mn†	15	0.950	3.940	1.280	2.000	2.400	3.020	1.680	2.200
RL‡ (Bq/l)	800	384.64	619.93	316.58	295.23	208.11	229.61	149.24	137.25
^{226}Ra	0.2	0.061	0.068	0.061	0.078	0.049	0.062	0.042	0.085
U_{nat}	0.05	0.003	0.014	0.003	0.004	0.005	0.005	0.010	0.014
Cu	0.1	0.014	0.117	0.008	<0.002	0.010	0.004	0.036	0.077
Hg	0.0005	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Ag	0.02	0.051	0.055	0.056	0.018	0.009	0.004	0.031	0.031
Pb	0.05	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004
As	0.05	0.004	0.015	0.001	<0.001	0.002	0.002	0.001	<0.001
Se	0.05	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
V	0.05	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Cd	0.01	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003
Cr	0.2**	0.006	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Fe	2**	0.060	0.130	0.130	0.050	0.200	0.120	0.250	0.850
Mn	0.3**	0.010	0.250	0.010	0.010	0.010	0.010	0.110	0.250
NH_4^+	0.5**	0.040	0.120	nd	0.010	0.040	0.240	0.040	0.070
NO_2^-	0.1**	nd	0.010	nd	0.020	nd	0.020	nd	<0.01
NO_3^-	50**	22.420	10.230	nd	16.670	nd	8.370	nd	8.350
SO_4^{2-}	250**	77.79	209.35	69.97	39.92	54.74	48.32	45.68	39.10

* Biological consumption of oxygen in 5 days

† Chemical consumption of oxygen determined by Mn

‡ Dissolved matter

** Slovak Technical Norm No. 75 71 11 Drinking Water

A "HERITAGE" FROM URANIUM MINING IN POLAND

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1. Introduction

In the Lower Silesia region of Poland, extensive uranium exploration and mining activities took place in the early 1950s under the supervision of Soviet engineers and managers. Uranium mining operations were opened at Kowary, Podgórze, Radoniów, and Kletno sites, while extensive uranium exploration was developed in numerous localities over Lower Silesia. Ore from three underground mines was transported directly to the Soviet Union for twelve years (1951-1963). Chemical processing of low-grade ore waste in Kowary began in 1969 and lasted until 1972.

Chemical processing of uranium ore required the construction of tailings ponds, one at Kowary. Both mining and processing of uranium ores in Poland ended more than 25 years ago. The companies responsible for the related environmental problems no longer exist. Nevertheless, the need to remediate the environment is real.

2. Legal aspects

There are no laws and regulation in Poland specifically directed toward the remediation of uranium mining areas. However, the following legislation on related issues is applicable for uranium remediation activities and should be taken into consideration:

- Geological and Mining Law,
- Law for the Protection of Agricultural and Forestry Land,
- Atomic Law,
- Regulation concerning Ionizing Radiation (National Atomic Energy Agency, NAEA),

- Regulation concerning Radioactive Wastes (NAEA),
- Regulation Concerning Permissions for Activities involving Ionizing Radiation (NAEA),
- Regulation Concerning Water Classification (Ministry for Environmental Protection, Natural Resources and Forestry),
- Law Concerning the State Inspectorate for Environmental Protection,
- Civil Engineering Law, and
- Law for the Protection and Forming of the Environment.

The Geological and Mining Law stipulates that the state is responsible for the liabilities from uranium production in Poland that ended in 1972. Therefore, the government must secure the funding for remediation measures, either through the national or district Environmental Protection Fund.

Polish regulation specifies no maximum allowable concentrations for natural radioactive isotopes, with a few exceptions. Limits for chemically hazardous contaminants are partially available from several regulations.

The regional authority (Voiodship) and its special services or offices are responsible for particular aspects of the remediation, e.g. civil engineering/building. The regional authority approves remediation plans and supervises the remediation. Inspectorates of Environmental Protection of a Voivodship—subordinated to the State Inspectorate of Environmental Protection—are responsible for environmental monitoring, while the President of the National Atomic Energy Agency is responsible for radiological monitoring as a part of the environmental monitoring.

3. General Description of Tailings Pond

The Town of Kowary was the center of uranium mining activities in Poland. The headquarters of the uranium mining company ZPR-1 (Zaklady Przemysowe R-1) were located there. ZPR-1 was the only uranium processing plant in Poland.

Mining of uranium in Poland ended in 1963, but hydrometallurgical processing of low-grade waste ores was continued at Kowary until 1972. As a result of these processing activities, a significant volume of solid wastes was produced and stored in the tailings pond.

The tailings pond in Kowary is situated in the north-south trending Jedlica valley. The Karkonosze Mountains form the western and southern slopes of this valley, while the Rudawy Janowickie Mountains form its eastern boundary. Jedlica Spring originates below the Okraj Pass, from which the Jedlica River flows toward the northwest and then directly north. The Jedlica River is a typical mountain stream with frequent changes in its water level.

The body of the pond is situated on the right (eastern) bank of the Jedlica River on the western slope of 853.3-m high Rudnik Mountain. It is enclosed on three sides by a dam which has been modified three times. The pond has an isometric oval shape and, since the valley gently slopes north, the northern dam is the highest. Its maximum

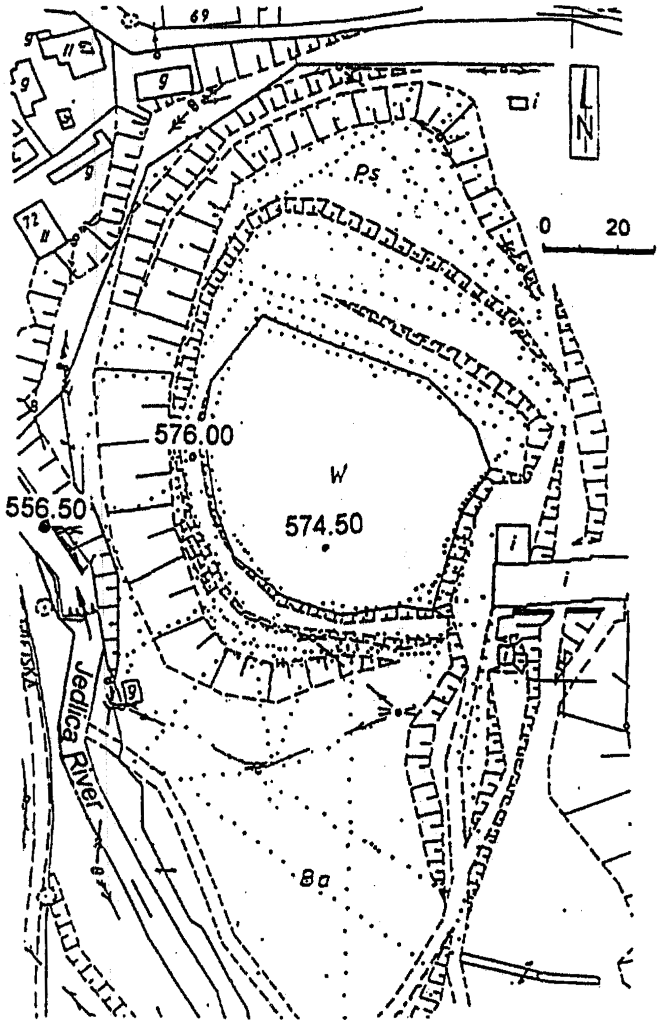

Figure 1. Plan view of the Kowary tailings pond.

height reaches 12 m at the northwestern corner, located close to the axis of the valley. The surface area of the tailings pond is 13,000 m^3 and the base area is 20,000 m^3.

About 250,000 tons of pulverized gneiss (grain size of order 0.1 mm), containing approximately. 30 ppm of uranium and from 0.03 to 0.08 ppm of radium, has been stored in the pond. The levels of ionizing radiation in pond and its vicinity are shown in Fig.2 [2].

Monitoring of ionizing radiation has been started. Monitoring equipment was provided by PHARE Project PH4.02/94 "Preparing Remediation at Uranium Milling and Mining Sites in the Phare Countries. Provision of Means to Assess Radiological Risks."

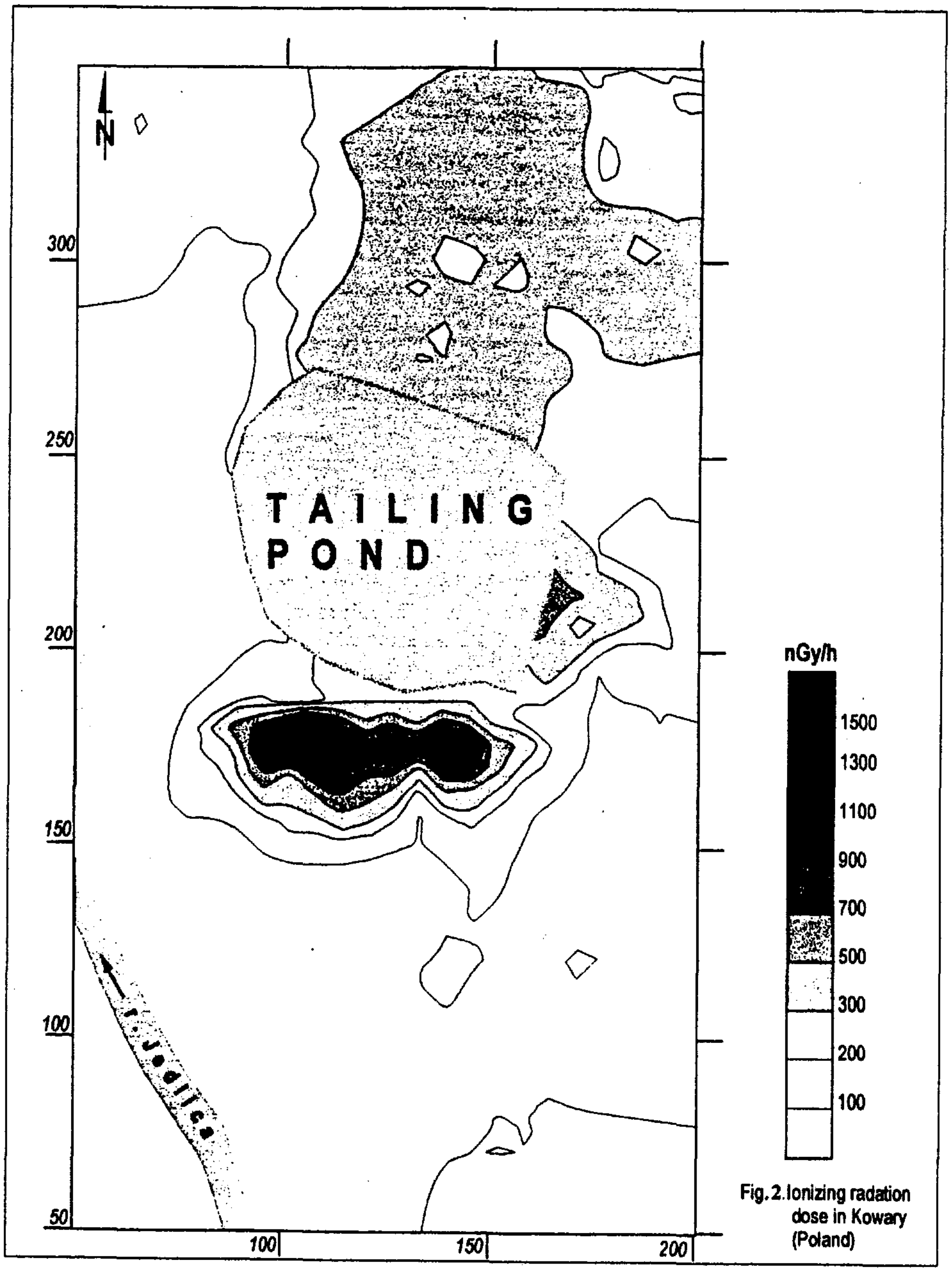

FIGURE 2. Ionizind radiation dose at Kowary tailings pond

In the early seventies, Wroclaw University of Technology (WUT) received, by governmental decision, ownership of the land and facilities of the former uranium company ZPR-1. The company Hydromet Ltd., owned by WUT, has continued to use the existing chemical plant for development of processes on nonferrous, rare (radioactive) metals, chemical production, and electrochemical processes. As a result, about 300 t of wastes from nonferrous and rare metals processing and about 500 m^3 of electrochemical processing fluids with up to 30 t of high solids content (Al, Ni, Zn and Na sulfates) have also been disposed in the pond.

4. Remediation programme

Prompt remedial action is necessary because summer rains cause heavy erosion in the steep mountainous valley in which the tailings pond is located. Buildings in the town of Kowary are located close to the 12-m-high dam. The Jedlica River, which is subject to flash flooding, flows along the foot of the western dam. Average annual rainfall at the altitude of the pond is about 750 mm, but it increases with altitude, reaching an average value of 997 mm at the top of Karkonosze. During the summer of 1997, after a daily rainfall of about 100 mm, flood waters of the Jedlica River eroded the western part of the dam.

Since 1996 Poland has participated in the PHARE Multi-Country Environmental Sector Programme "Remediation Concepts for Uranium Mining Operations in CEEC" [1]. The objectives of this Programme are:

- to inventory the existing situation,
- to implement a pilot project,
- to support co-operation between interested countries.

Within the Programme, an inventory and one common data base for the CEEC have been developed. According to this inventory, the situation in Poland is characterised by a large number of small-scale liabilities from uranium exploration, distributed over several locations in the country and generally having minor impact on the environment. Only a limited number of sites related to mining and milling have a more serious impact. The most important of them is tailings pond in Kowary

The objectives of the remediation programme for the tailings pond in Kowary designed by Wroclaw University of Technology (WUT) are as follows:

- hydrogeological, hydrological, and environmental mapping of the pond and its surroundings,
- chemical, radiochemical, and bacteriological characteristics of the environment at the pond,
- design for remediation and monitoring of the pond,
- construction of a water drainage system,
- construction of a water treatment plant,
- dewatering of the pond,
- reclamation of the pond and its surroundings,

- continuous radiochemical and chemical monitoring of air, soil, and seepage waters as well as monitoring of dam's stability.

The financing of this program is expected from the European Commission (Directorate General XI) [3], regional and national Environment Protection Funds, and WUT.

5. Conclusions

Completing the remediation of the tailings pond at Kowary would remove the ecological threat originating from former uranium mining/processing operations. Its localization and size makes this particular remediation project both specific and possible to complete within a short period of time. This is quite realistic since several preliminary investigations, measurements, and studies necessary for planning and designing the remediation have been completed by WUT under financing by both WUT and regional (Jelenia Góra) Environment Protection Fund.

6. References

1. PHARE Pilot Project Proposal. Remediation of Tailing Pond in Kowary, WUT, Wroclaw 1977.
2. Environmental Impact of the Tailing Pond in Kowary, WUT, Wroclaw 1998.
3. Remediation of Tailing Pond in Kowary. Application to EC DG XI, WUT, Wroclaw 1998.

A FIELD STUDY OF CAPILLARY AND HYDRAULIC BARRIERS IN LANDFILL COVERS VARYING IN SLOPE AT LOS ALAMOS, NEW MEXICO, USA

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1. Introduction

The tailings pond at Sillamäe, Estonia, is an oval retention impoundment on the waterfront of the Gulf of Finland, with an overall area of about 330,000 m^2 [1]. The top of the dam is about 25 m above sea level and a tailings pond containing about 150,000 m^3 of acid wastewater covers about 30% of the area. The tailings pond contains about 8 million m^3 of wastes, including uranium tailings, oil-shale fly ash, and loparite ore processing tailings. Environmental concerns include the stability of the dike, leaching of uranium and toxic heavy metals into the ground water and into the Gulf of Finland, and radon and dust emissions from the wastes.

The goal of disposing of radioactive and hazardous wastes such as those at Sillamäe using shallow land burial is to reduce risk to human health and to the environment by isolating contaminants until they no longer pose a hazard. VBB studies suggest that about half of the annual precipitation infiltrates the Ranstad tailings cover near Skovde, Sweden [2], and if similar water balance relationships occur in Sillamäe, the proposed engineered cover placed over the tailings pond will certainly be challenged in terms of performance. In the USA, institutional control and maintenance of low-level radioactive-waste repositories are expected to cease 100 years after the closure of a waste site. After this time, the repository's engineered barriers and geohydrologic conditions need to act passively to isolate the radionuclides for an additional 300 to 500 years [3]. Even though the successful performance of the entire landfill is very much a function of interactive water balance processes [4], traditional remedial engineering solutions have ignored these processes, leading to numerous landfill failures [5, 6]. Field water balance data for landfill cover designs do not exist to enable the site operator to adequately define and engineer suitable barriers to prevent the migration of waste materials out of the landfill.

2. Development of Technology for Shallow Land Burial

A field research program was initiated at the Los Alamos National Laboratory in 1980 to develop technology for the closure and stabilization of shallow land burial sites [7, 8]. The environmental distribution and migration of transuranics, cesium, strontium, and tritium was evaluated in the field in 6-m-deep caissons with a diameter of 3 m at the Los Alamos Experimental Engineered Test Facility (EETF; ref. 9) and at material disposal areas at Los Alamos [10-14]. Various factors influencing these contaminant distributions were evaluated as being part of an interdependent the shallow land burial system, as opposed to a traditional engineering approach [12, 15-20]. Since the earliest studies in this program suggested that man-made materials such as asphalt, hypalon, and concrete have a field life of no more than 25 years [21], cobble, gravel, and clay were evaluated at the EETF and at material disposal areas as a component of the landfill cover to limit the intrusion of both roots of clover, alfalfa, and several grasses and burrowing pocket gophers [17, 22-24]. These studies were performed in conjunction with laboratory and field studies of susidence [25, 26] and geotechnical studies of our local volcanic tuff backfill and mixtures of this and bentonite [27].

Soil erosion studies of simulated landfill covers were performed on standard-sized field plots (3.05 by 10.7 m) using rainfall simulator technology developed by the US Department of Agriculture [28-31]. Instrumentation used to support these and other field studies for the measurement of soil water tension and volumetric water content were evaluated [32-36], as well as methods for sampling soil water in unsaturated soils [37]. The soil erosion studies then evolved into more comprehensive studies of various landfill cover designs and cover treatments where all of the elements in the water balance equation were measured in the field [23, 38-43], such as in the current field study. Modeling studies of water balance relationships in landfill covers and of contaminant migration [44-48] were also performed. With the help of the US Department of Energy's International Technology Exchange Program, these studies also directly involved current European research [49-63].

3. Protective Barrier Landfill Cover Demonstration

The shallow land burial studies described in the previous section were used to design the Protective Barrier Landfill Cover Demonstration at the Los Alamos National Laboratory in Los Alamos, New Mexico. The objectives of the present study are: (i) to determine if hydraulic and capillary barriers in three landfill cover designs can change water balance relationships over those observed in landfill covers without engineered barriers; and (ii) to determine how the slope of the landfill cover influences water balance parameters.

4. Materials and Methods

4.1 PLOT CONSTRUCTION, DESIGN AND RATIONALE

The Protective Barrier Landfill Cover Demonstration was constructed to compare water balance on the conventional landfill cover design, similar to that used in Los Alamos and by the waste management industry for waste disposal [5], with that on three other designs containing engineered barriers (Fig. 1). The performance of all four designs was evaluated at dominant downhill slopes of 5, 10, 15 and 25% on plots without vegetation. These 16 plots were installed in 1991 in our 8-ha field test facility [9] and were instrumented so that a complete accounting of precipitation falling on the plots could be measured. The plots were constructed and instrumented to provide measures of runoff and interflow, as well as seepage and soil water storage as a function of slope length.

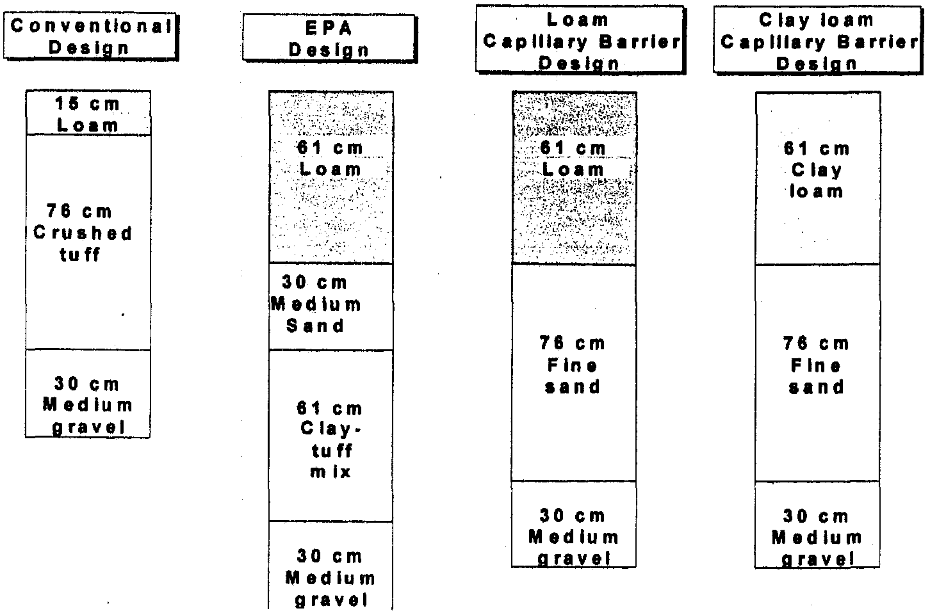

FIGURE 1. Description of soil layers in the four landfill cover designs at the Protective Barrier Landfill Cover Demonstration. A high conductivity geotextile was installed above the medium gravel layer and the medium sand layer.

The Protective Barrier Landfill Cover Demonstration was emplaced on an east-facing slope similar to the aspect of many of the local landfills where this technology will be applied. The area was surveyed into four pads, each of which received crushed tuff to establish the varying downhill slopes. Four 1.0- by 10.0-m plots were then constructed on each pad [41]. A seepage collection system was installed in the bottom of each of the plots consisting of four metal pans filled with medium gravel (8.0- to 25-mm diam) overlain with a high conductivity MIRAFI geotextile used in previous field studies [39]; an 11-cm-wide space was left between the sidewalls of the plot and the pan to minimize sidewall effects.

The hydrologic properties of soils used in the field study are presented in Table 1. The soils were analyzed for porosity and for hanging column and thermocouple psychrometric moisture retention characteristics [64]. Constant head determinations of saturated hydraulic conductivity were performed as well as pressure plate extractor determinations of moisture retention characteristics [65]. Van Genuchten's RETC code [66] was employed to determine the van Genuchten factors for each soil using analytical procedures described previously [67, 68].

TABLE 1. Hydrologic properties of soils used in field study as determined with van Genuchten's RETC model [66] and laboratory analyses.

	van Genuchten factors			θ_r	θ_s	Saturated Conductivity
Soil Description	α	n	m	(cm^3/cm^3)		(cm/s)
Loam topsoil	0.0271	1.539	0.3504	0.0692	0.4209	5.7×10^{-3}
Hackroy clay loam	0.0100	1.548	0.3541	0.0730	0.4839	2.5×10^{-4}
Fine sand	0.0334	5.472	0.8173	0.0700*	0.4180	1.2×10^{-2}
Medium sand	0.0288	3.766	0.7344	0.0376	0.4184	1.3×10^{-1}
Crushed tuff	0.0104	1.707	0.4140	0.0031	0.4079	8.2×10^{-4}
Clay-tuff mix	0.00014	3.992	0.7495	0.0000*	0.4415	6.3×10^{-8}
Medium gravel	-	-	-	-	-	2.0

* Constrained parameter in van Genuchten model.

The technology for controlling soil water erosion on all cover designs consisted of applying a 70% ground cover of medium gravel (8.0- to 25-mm diam). The plots with the Conventional Design, similar to that used at Los Alamos waste sites, contained 15 cm of a loam topsoil (Fig. 1) consisting of a 2:1:1 (V:V:V) mixture of an uncharacterized topsoil, sand, and aged sawdust (<9.5-mm diam). This topsoil was not underlain by an engineered barrier, only with 76 cm of crushed [12, 39].

One set of plots contained the EPA-recommended [69] final cover design (Fig. 1). These plots contained 61-cm of the loam topsoil described previously emplaced on top of 30 cm of medium sand (0.25 to 0.5-mm diam). The medium sand layer corresponds to the EPA "drainage layer" and was overlain with the MIRAFI geotextile to provide the EPA-recommended filter layer necessary to prevent fine soil particles from migrating into the drainage layer. The bottom layer in the EPA-recommended final cover, called the "low-permeability layer," usually consists of a 20-mil (0.5 mm) minimum thickness flexible membrane liner (FML) on top of a 60-cm-thick layer of soil with an in-place saturated hydraulic conductivity of $<1\times10^{-7}$ cm/s. Since the plastic FML would last less than 35 years [69], this feature of the EPA design was omitted in our EPA Design. The results of previous research on mixtures of local crushed tuff and sodium-saturated bentonite [25, 27] indicated that a 1:10 (W:W) dry mixture of finely ground Aquagel (Baroid Drilling Fluids, Farmington, NM) and crushed tuff (called the clay-tuff mixture) should easily provide the low saturated hydraulic conductivity required for this layer (Table 1).

Two designs contained capillary barriers varying only in the type of topsoil (Fig. 1). One of the designs contained 61 cm of the loam topsoil used in the previous designs, whereas the other design contained 61 cm of a Hackroy clay loam classified as a Lithic Aridic Haplustalf (clayey, mixed, mesic family) and used in two previous studies (Nyhan et al., 1984, 1990a). These soils were emplaced on top of 76 cm of a fine sand (0.05-to 0.425-mm diam) made in the sand classifier/blender. The fine sand was specifically chosen to complement the underlying medium-sized gravel in terms of optimizing the relationship between the hydraulic conductivity and the water-holding properties of the capillary barrier [63].

4.2 MEASUREMENT OF SEEPAGE, INTERFLOW, RUNOFF, AND PRECIPITATION

Runoff, precipitation, and seepage were collected year-round from December 1991 through July 1995, as well as interflow (flow occurring along the length of each plot through the medium sand layer in the EPA Design, the fine sand layer in the two designs with capillary barriers, and the crushed tuff layer of the Conventional Design). Water levels in each 100-liter tank used to collect these data were measured with a microprocessor-controlled ultrasonic liquid level sensor (model DCU-7, Lundahl Instruments, Logan, UT) connected to a multiplexed, automated system described previously [41]. The water levels in the tanks were routinely recorded hourly, but much more frequently when the tank was either emptying or when it was nearly full.

Precipitation was measured using a weighing rain gauge and a long-term event recorder.

4.3 MEASUREMENT OF SOIL WATER CONTENT

Soil water content was routinely monitored once every six hours from December 1991 through July 1995, at each of 212 locations throughout the 16 plots using Time Domain Reflectometry (TDR) techniques with the help of an automated and multiplexed measurement system. Volumetric water content was measured with a pair of stainless steel waveguides (60-cm long, 3-mm diam soil moisture probes; model number 6860, Campbell Scientific, Logan, UT), which are buried parallel and 5 cm apart in the soil and are connected to a 26-m length of RG-8/U coaxial cable. TDR waveguides were emplaced in the Conventional Design at depths of 5-10, 20-80, and 80-86 cm, in the EPA Design at depths of 1-61, 61-91, 96-102, and 92-152 cm, and in the two designs containing capillary barriers at depths of 1-61, 66-126, and 126-132 cm. These TDR waveguides were normally emplaced at downslope locations of 2.63, 4.65, 6.62, and 8.69 m for each soil depth, except at the deepest depths in the Conventional Design and the designs containing the capillary barriers, where they were emplaced at downslope locations of 3.64, 5.66, 7.68 and 9.70 m (to coincide with the bottom end of each of the four seepage pans installed in the bottom of each field plot).

4.4 WATER BALANCE CALCULATIONS

Daily water balance calculations were performed by determining the daily change in soil water inventory, by summing the daily amounts of precipitation, seepage, interflow, and runoff, and then determining the amount of daily evaporation by difference. As an independent check on these evaporation estimates, evaporation was also estimated from eddy heat flux data collected from a fast-response hygrometer mounted at a height of 12 m on a 92-m meteorological tower at Los Alamos; daily values were estimated from field data collected at 15-minute intervals.

In order to further evaluate the water balance data, daily short-wave radiative energy received by field plots with slopes of 5, 10, 15, and 25% was estimated from pyranometer data collected at a height of 1.2 m from the same meteorological tower described above at the same sampling frequencies. The influences of slope and seasonality of short-wave radiative energy were calculated using the SOLARFLUX model [70].

5. Results and Discussion

5.1 ESTIMATES OF PRECIPITATION AND SOIL WATER INVENTORY

The overall significance of each year's water balance data can best be explained by understanding the spatial and temporal occurrence of precipitation around Los Alamos [71]. Bowen showed that mean annual precipitation is 32.8 cm at White Rock, the only station close to the Protective Barrier Landfill Cover Demonstration with a data base longer than the data collected in this field study. We determined that 2.94-year, 5.56-year, and 20-year events occurred in 1992, 1993, and 1994, respectively.

Soil water inventory data are presented for several layers of the Conventional Design (Fig. 2). The inventory data for the loam topsoil represents the daily average readings of horizontally placed waveguide pairs at a depth of 5 to 10 cm at downslope locations of 2.63, 4.65, 6.62, and 8.69 m. The frequent variations in the TDR measurements at this depth occurred because soil water content usually increased with precipitation events as small as 0.5 cm. Similar data collected at the 15-75 cm depths exhibited less frequent fluctuations because the small precipitation events did not penetrate to the maximum depths over which the measurements were integrated.

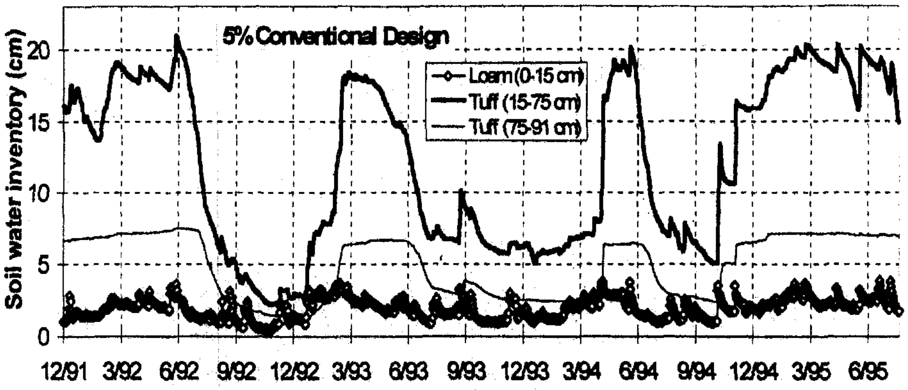

FIGURE 2. Daily soil water inventory as a function of time for the Conventional Landfill Cover Design with the dominant downhill slope of 5%.

Large changes in soil water inventory were observed at both sampling depths in the crushed tuff layer monitored with time in the Conventional Design with the 5% slope(Fig. 2), typical of the changes observed on the three field plots containing this design with larger slopes. The soil water inventories presented for the 15-75 cm depth decrease throughout the summer and fall of each year and increase during the cooler winter and spring months with snowmelt additions. The data collected at the 75-91 cm depth of the tuff layer (Fig. 2) shows that soil water inventories remained at values greater than 2.9 cm (corresponding to field capacity volumetric water content of 18%) over 69% of the time. Since soil water inventory values greater than 2.9 cm for the tuff correspond to soil water regimes dominated by gravity flow, these time periods represent periods when seepage was observed to occur beneath this lower tuff layer with additions of water from upper layers.

5.2 WATER BALANCE SUMMARIES

As might have been expected in a semiarid environment, 92 to 97% of the precipitation received by all of the landfill cover designs was evaporated from these unvegetated landfill cover designs. Although the soil in the vicinity of the meteorological tower is slightly heavier-textured than the loam topsoil in the Conventional Landfill Cover Design with the 5% slope, we were able to compare the tower hygrometer estimates of water flux with the amounts of evaporation observed in this plot. We discovered that the eddy heat flux data from the meteorological tower (collected from December 1991 through July 1995) amounted to 131.1 cm water, compared with our field plot estimate of 159.5 cm water for the same time period.

Evaporation usually increased with increases in slope within each landfill cover design (Table 2) on our east-facing study site, because plots with large slopes intercepted more short-wave radiative energy than plots with smaller slopes (Fig. 3). This effect was dominant during the first and fourth quarters of each year, during

TABLE 2. Water balance data for all landfill cover designs from December 1, 1991 through December 31, 1997. Total average precipitation received by all 16 plots for this time period was 276.1 cm.

	Water balance parameter (cm)				
Design and Slope (%)	**Evaporation**	**Interflow**	**Seepage**	**Runoff**	**Change in soil water inventory**
Conventional design					
5%	260.0	10.6	2.8	3.6	2.7
10%	270.0	14.8	1.3	6.5	-12.0
15%	271.2	9.8	1.3	4.9	-7.3
25%	271.8	7.8	0.5	7.2	-8.2
EPA design					
5%	256.5	21.6	0.0	2.6	-5.2
10%	254.0	21.6	0.0	2.3	-4.8
15%	257.5	18.5	0.0	5.4	-6.1
25%	253.9	17.8	0.0	8.2	-4.6
Loam capillary barrier design					
5%	257.7	17.4	1.2	1.4	-3.5
10%	245.6	24.2	0.6	5.7	-2.2
15%	255.8	21.0	0.0	5.0	-4.9
25%	260.7	12.5	0.0	7.7	-6.1
Clay loam capillary barrier design					
5%	255.4	15.9	0.7	3.2	-0.1
10%	255.5	14.2	0.0	7.1	-0.6
15%	258.7	7.6	0.0	8.9	-0.8
25%	264.0	1.4	0.0	9.9	-2.4

times when seepage occurred; i.e., during the fourth quarter of 1993, plots with a 5% slope received 1339 MJ/m^2 short-wave radiative energy compared with the 1561 MJ/m^2 received by the plots with a slope of 25% (Fig. 3). Consequently, the sum of the interflow and seepage usually decreased with increasing slope for each landfill cover design (Table 2). Slightly larger amounts of evaporation occurred on the plots with the Conventional Design than on those with the Loam Capillary Barrier Design. A partial explanation for this observation is that soil water was removed faster through interflow and seepage in the underlying fine sand layer in the Loam Capillary Barrier Design than through the crushed tuff layer in the Conventional Design, considering the respective saturated hydraulic conductivity values were 1.2 x 10^{-2} and 8.2 x 10^{-4} cm/s (Table 1).

Although runoff did not seem to be related to surface slope on a per event basis, runoff did increase with increasing slope over the 44-month duration of this study for each of the designs. Runoff generally accounted for about 1-4% of the precipitation losses across all of the plots studied (Table 2).

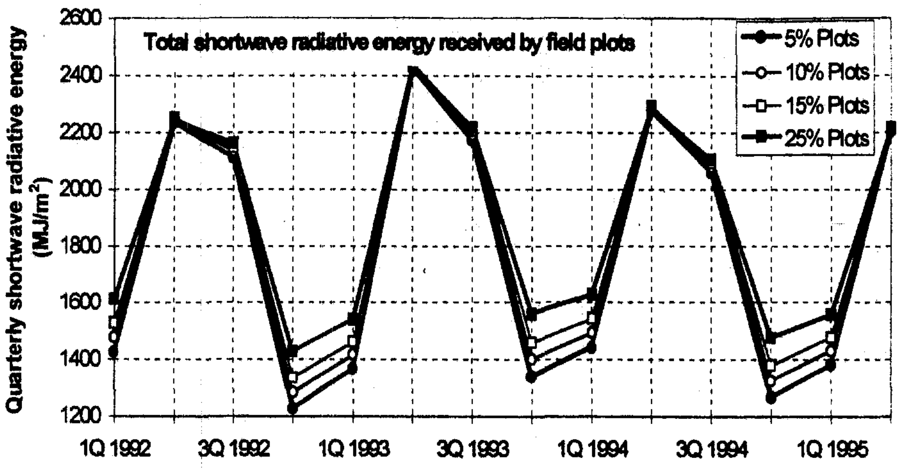

FIGURE 3. Quarterly total short-wave radiative energy received by field plots with slopes ranging from 5 to 25%. Data were estimated from 15-minute meteorological tower observations that were corrected for slope using the SOLARFLUX model (Rich et al., 1995).

The site operator usually prefers minimal seepage to occur on the landfill. Seepage was definitely decreased with engineered barriers in this study over that observed in the Conventional Design, which did not contain an engineered barrier. The Conventional Design with the 5% slope exhibited 10.6 cm of interflow and 2.8 cm of seepage from 1991 through 1997, representing a total of 13.4 cm of water that would migrate through this profile in a waste trench at Los Alamos not containing the gravel layer in the Conventional Design (Fig. 1). In contrast, the hydraulic barrier in the EPA Design effectively controlled seepage at all slopes, and both of the capillary designs worked effectively to eliminate seepage at the higher slopes (Table 2), many of which are commonly used on waste sites at Los Alamos and throughout the waste management community.

5.3 COMPARISONS WITH SILLAMÄE AND CONCLUSIONS

We are currently learning more about the Sillamäe tailings pond, but a few observations can be made about the difference in climate between Los Alamos, NM and data collected at Tallinn (Figs 4, 5). The distribution of average temperature throughout the year is similar for Los Alamos and Tallinn. However, the average annual temperatures at Tallinn and Los Alamos are about 5.5 and 8.8 C, respectively, and the range in the average maximum and minimum monthly temperatures is consistently much larger for Los Alamos (13.3 C) than for Tallinn (5.7 C).

Tallinn receives about 560 mm of annual precipitation, whereas the annual precipitation in Los Alamos is about 468 mm. Most of Los Alamos's precipitation

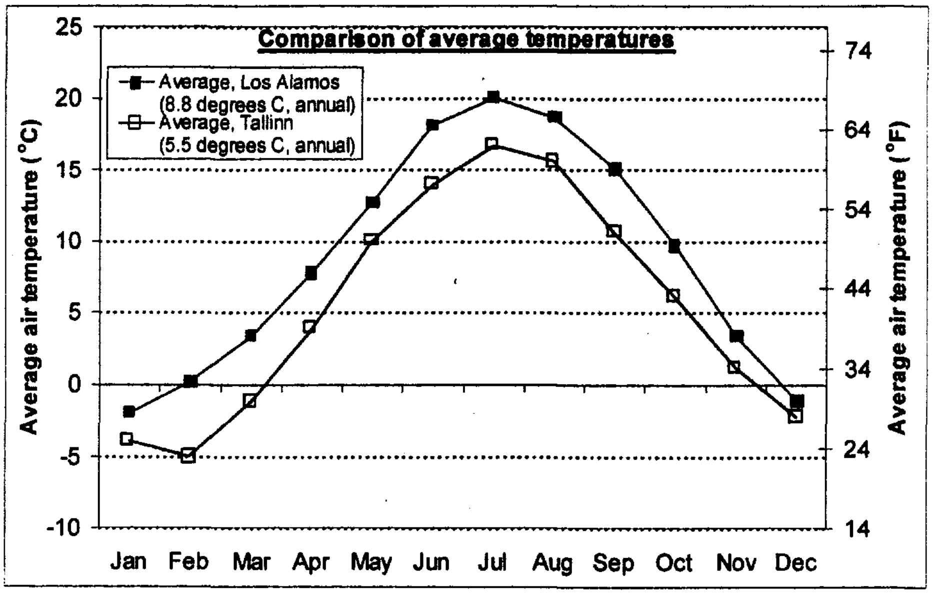

FIGURE 4. Comparison of average temperatures at Los Alamos and Tallinn.

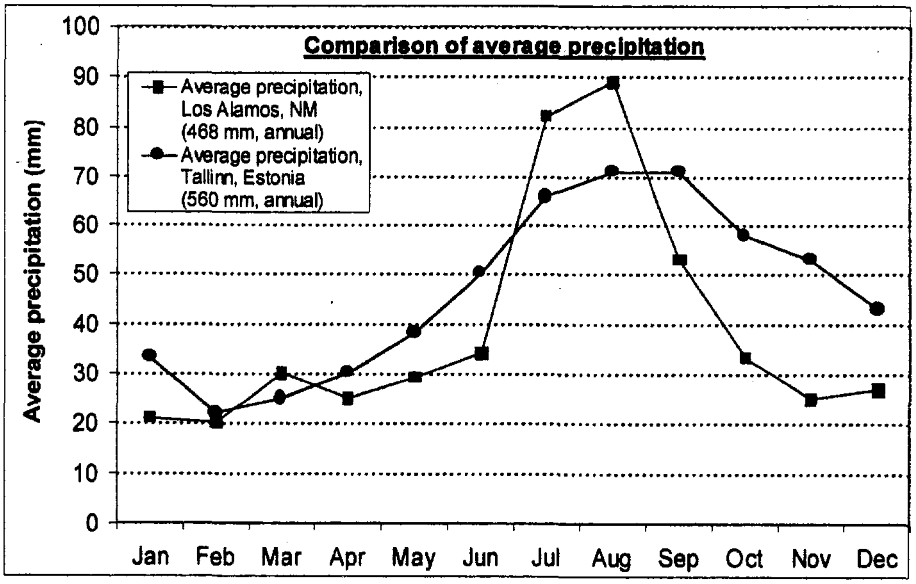

FIGURE 5. Comparison of average precipitation at Los Alamos and Tallinn.

occurs as intense rains in July and August with much smaller amounts of precipitation occurring throughout the rest of the year. Tallinn's precipitation pattern exhibits a similar pattern, but with smaller differences between precipitation received between July through September and the rest of the year. However, Tallinn receives more precipitation than Los Alamos does during time periods of low solar radiation, a factor that will enhance potential seepage through shallow land burial facilities.

Current state and federal regulations in the USA usually require an engineered barrier to be present in the landfill cover design, a design criterion that is also impacted by risk assessments and cost considerations. Capillary barriers can be used as alternative designs to the EPA Design [69], with the realization that seepage did occur at 5 and 10% slopes in the Loam Capillary Barrier Design and in the Clay Loam Capillary Barrier Design with a slope of 5% (Table 2). Although the EPA Design does seem to eliminate seepage in field plots with 5 and 10% slopes (Table 2), the EPA design is probably more expensive than alternative designs [4]. In the case of either engineered barrier, other field data sets similar to that collected in the current study are needed in a variety of climates and with slope lengths longer than 10 m to validate hydrologic models that can be used in design selection.

6. References

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ESTONIAN RADIATION PROTECTION REQUIREMENTS AND THE SILLAMÄE SITE

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The Estonian national radiation protection and safety infrastructure is now being developed. Comparatively rapid changes in the establishment of the legal framework, as well as the regulatory components of the infrastructure, are in progress. The existing legislation enacts the concepts, principles, and criteria for efficient regulation of radiation safety in Estonia. Special articles regulating the exposure of workers and the general population to enhanced natural radiation are included both in the Radiation Act and the supporting legislation. A specific procedure is enforced for deciding whether work activities involving significant natural exposure should be considered as radiation practices. A brief overview of the requirements relevant to the Sillamäe site, together with measurement and assessment data, is presented.

1. Overview of Legislative Acts

1.1 THE RADIATION ACT

The Radiation Act (*Kiirgusseadus*) [1] as the principal legal instrument of the radiation protection infrastructure was brought into force only in 1997. The Act is based on the concepts, principles, terms, and limits laid down in the International Basic Safety Standards (BSS) [2] and Directive 96/29/EURATOM [3]. The basic internationally approved principles—justification of practices, optimisation of protection and safety, limitation of individual doses, adoption of justified and optimised interventions, the primary responsibility of the legal person (licensee), and authorisation 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 also adopted.

The Act specifies the dose limits of occupational exposure for radiation workers, apprentices, students, and pregnant women and those of exposure for the general public in accordance with those of the BSS.

An independent competent authority, the Estonian Radiation Protection Centre (ERPC, *Eesti Kiirguskeskus*), is empowered to authorise practices by licensing, to inspect 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 and to ensure supervision. The ERPC, administered by the Ministry of Environment, began to fulfil its duties in 1996.

The Act consists of 36 paragraphs in 6 chapters. A brief review of the items covered in the Act follows.

Chapter 1, General Provisions, includes the purpose and scope of the Act, terms and principles for acceptable practices and interventions, and authorisation of the ERPC for the enforcement of the requirements.

Chapter 2, Requirements for Practices, includes licence requirements and exemptions, obligations of the licensees, and cancellation of licences.

Chapter 3, Exposures, includes dose limits for occupational and public exposure, requirements for natural exposure, obligations of workers, dose register, age limit, medical surveillance of workers, assessment of public exposures, and requirements for medical exposure and emergency exposure. Dose limits as the basic regulative criteria are explicitly included in the Act (Table 1).

TABLE 1. Dose limits for radiation workers and population

Doses	**Occupational exposure (mSv/a)**	**Training of apprentices (mSv/a)**	**Public exposure (mSv/a)**
Effective dose			
5 year average	20	6	1
any single year	50		5
Equivalent dose			
lens of the eye	150	50	15
skin and extremities	500	150	50

Chapter 4, Radiation Sources, includes the safety of sources, installation and repair of sources, type approval, and transportation of radioactive substances and wastes.

Chapter 5, Radioactive Wastes, includes management principles and requirements, transfer and restrictions, export requirements, and an import prohibition for disposal.

Chapter 6, Final Provisions, includes supervision, implementation of the Act, and sanctions for legal persons for breach of the provisions.

Considering the extensive use of international and EU legislation as models in drafting the Act, a good accordance between the provisions of the Act and the model ones is not surprising. A recent analysis showed a full or partial harmonisation of its articles to the corresponding provisions of Directive 96/29/Euratom. In 1997 the competent experts of IAEA also pointed to a definite success in conversion of the requirements of [2,3] into national legislation. It should be noted that an amendment to the Act [25], concerning mainly its links to the requirements of the other recently enforced legal acts, was accepted by the Parliament.

Specifying the principal requirements for radiation protection and safety, the Radiation Act also empowers the Government and the Ministers of Environment and Social Affairs to issue decrees with specific regulations for its implementation. The procedure of the delegated legislation is in progress. Below we will review a few enforced and drafted decrees and other existing legal documents concerning radiation protection.

1.2. SUPPORTING LEGISLATION

The Decree on the order for issuance of licences for radiation practice [4] establishes detailed requirements for applicants and applications. The ERPC is authorised to assess all applications for permission to conduct radiation practices and, when the application is in compliance with the requirements, to issue a licence for radiation practice for a time period up to five years.

The decree on registration of radiation sources [5] regulates the process of registering radiation sources by the ERPC, establishes a national radiation source register, and introduces detailed requirements to the licensees. The source register is maintained by the ERPC.

The government decree on exemption levels for radiation sources [6] enforces exemption levels for radionuclide activity and specific activity. Terms and levels equal to those stipulated in the BSS and the EU Directive are incorporated. Exemption levels are considered basic criteria for decisions on licensing radiation practices. No licence is needed for operations with activities or specific activities of radionuclides below the exemption levels. Examples of the exemption levels relevant to the raw material and waste are given in Table 2.

TABLE 2. Exemption levels for radionuclides

Radionuclide	**Activity (Bq)**	**Specific activity (kBq/kg)**
Pb-210	10^4	10
Ra-226	10^4	10
Th-228	10^4	1
Th-230	10^4	1
Th-232 nat	10^3	1
U-235	10^4	10
U-238	10^4	10
U-238 nat	10^3	1

For multiple radionuclides or mixtures, the sum of their activity or specific activity ratios to the corresponding exemption levels should satisfy the following condition:

$$\sum_i \frac{A_i}{A_{t,i}} \leq 1 .$$

The government decree on safe transport of radioactive materials [7] enacts regulations for safe transport of radioactive materials, including radioactive wastes. The requirements of the BSS, the IAEA regulations and the EU directives are harmonised with existing local transport legislation to elaborate the regulations involved in the decree. The decree covers general provisions for radiation safety and emergency response, etc.; activity and fissile material limits; requirements for packages, marking, labelling, transport and storage in transit; test procedures; administrative requirements; documentation. Enactment might be expected in 1998.

The decree of the Minister of Environment on radioactive waste management [8] provides detailed requirements for radioactive waste management, including their storage, disposal, facilities, etc. The requirements of the recent IAEA RADWASS Safety Standards are included to the extent reasonable for the scope of practices with radioactive wastes in Estonia.

The Act on Export and Transit of Strategic Goods entered into force in 1994 [9]. It makes licences for the export and transit of these goods mandatory. Nuclear technology, equipment, special nuclear materials, nuclear waste, related materials, including metals and ores with Th or U concentrations higher than 0.05 %, are included in List 2 “Atomic technology and related materials and equipment” [10]. The list also includes dual-purpose materials: Ra-226 found in any compound and mixture (2.12), as well as radioactive compounds and isotopes (5.1.166). An Interdepartmental Commission issues the licences for export and transit of the listed goods.

The decree of the Minister of Environment on monitoring and evaluation of population doses [11] establishes the method of verification of population doses. In addition, radioactive releases are regulated in the following manner. In the case of natural radiation, a licensee for radiation practice or another undertaking shall arrange a special survey including monitoring and evaluation of population doses, if their activities or the circumstances involved , including radioactive releases or wastes, may cause enhanced population exposure. The following reference levels are established:

a) the annual collective effective dose for population > 1 *manSv*;
b) the annual individual effective dose > 10 *μSv*;
c) the specific activity index *I* of building materials or household water > 1.

The specific activity index *I* is calculated using the following expression:

$$I = \sum_i \frac{C_i}{A_i} \ ,$$

where Ci is the measured specific activity of the material (Bq kg-1), and Ai is the radionuclide-specific parameter (Tables 3 and 4). It should be noted that similar indices were adopted in Finland [10], which served as models for the Estonian decree.

TABLE 3. Parameters A_I for building materials

Material	Parameter A_I (Bq kg^{-1})			
	^{226}Ra	^{232}Th	^{40}K	^{137}C
Building material	300	200	3000	-
Materials for streets, playgrounds, etc.	700	500	8000	2000
Ground filling material	2000	1500	20000	5000

TABLE 4. Parameters A_i for household water

Material	Parameter A_i (Bq l^{-1}) ^{210}Pb	 ^{210}Po	 ^{226}Ra	 ^{228}Ra	 ^{234}U	 ^{238}U	 ^{222}Rn
Household water	0,5	3	3	2	20	20	300

The effective dose caused by radon/thoron progeny should be calculated using the following dose coefficients (Sv (J h m^{-3})$^{-1}$):

Radon ^{222}Rn	(homes)	1.1
Radon ^{222}Rn	(workplaces)	1.4
Thoron ^{220}Rn	(workplaces)	0.1

The adopted dose coefficients define the annual average concentration levels at workplaces, above which either remedial actions are needed or the work should be considered as radiation practice. E.g., for radon concentration 1000 Bq m^{-3} an annual effective dose of 6 mSv can be estimated at workplace (2000 *h*, equilibrium factor of 0.4). According to the EU directive [3], exposures of this magnitude are characteristic for A-category workers. The employer shall cover the expenses of the survey [1, § 14 (4)]. The reports of the surveys shall be presented to the ERPC for its expert opinion. The results of the surveys together with this expert opinion may form a basis for the decision by the Minister of Environment to regard the work as a radiation practice or otherwise [1, §14 (3)].

Detailed requirements for practices, radiation sources, etc., causing occupational exposure of workers are in preparation.

2. Implementation of the Radiation Act

The licensing procedure is the most important instrument for the implementation of the requirements of the Radiation Act. In the procedure, the EPRC should carefully examine the conditions and circumstances offered by the applicant of the licence regarding their compliance with the requirements of the Act and the supporting decrees. The licence for radiation practice or for transportation import and export of radioactive materials, etc., is granted if the EPRC decides that the applicant satisfies the requirements. However, the licensed radiation practice or other licensed activity remains under supervision by the ERPC. If the licensee does not comply with the requirements of the Act or the underlying documents or with the conditions of the licence, the licence may be revoked or withdrawn, its conditions may be changed, or the licensee may be fined.

2.1. PENAL SANCTIONS

Sanctions to legal persons concerning offences in connection with the misuse of radioactive materials and ionising radiation are included in the Radiation Act [1, Ch.6, § 36], while sanctions to persons concerning the above crimes are found in the Special Section of the Criminal Code of Estonia [13]. Chapter 2, "Offences against the state," contains a provision on illegal smuggling of radioactive materials. Chapter 11, "Offences against public order and social security," contains several provisions dealing with crimes in connection with illegal acquisition, storage, use and transfer of radioactive and nuclear material (§ 208^3, § 208^4) or with the threatening of acquisition and misuse of these materials (§ 208^5). §208^6 penalises persons, who in violation of the provisions for storage, use, registration, transport and handling of these materials, cause personal injury, death or other major damage or the risk of those mentioned.

It should be mentioned that even the maximum fines for legal persons (up to USD 2200) might appear rather marginal. However, the Act also enforces the obligation of the supervisor to suspend the practice violating the provisions of the Act for up to six months and to prescribe the termination of breach [1, § 9 (1)].

3. Exposures at Sillamäe

Due to past and present work activities and the types of raw material used in production at the Sillamäe plant, the radiation safety issues are or may be of concern for both occupational and public exposure. At the same time, unfortunately almost no public data is available on the exposures to workers involved in activities at the site. The radiological situation outside the premises has been studied in more detail. However, the complex nature of the natural background radiation sources in the region (e.g., high and varying ^{226}Ra/^{238}U concentrations in soil, high outdoor ^{222}Rn levels, etc.) [17-21] greatly interferes with reliable assessments of the technological impact on public exposure.

Below a few circumstances specific to Sillamäe, together with their relevance to the corresponding legal requirements, will be briefly discussed.

3.1. RAW MATERIAL AND WASTE

It is known (see, e.g., [14]) that the production of rare-earth metals involves processing of ores with high concentrations of uranium (up to 10 kBq kg^{-1}) and thorium (up to 100 kBq kg^{-1}), while in waste streams and dust the concentrations may be even higher. In niobium production, activity concentrations of thorium reaching 50 kBq kg^{-1} have also been found for columbite. Since the beginning of the 1970s, loparite has been extensively processed for production of niobium, tantalum and rare-earths at Sillamäe. Loparite contains as an average about 4 kBq kg^{-1} ^{238}U and 25 kBq kg^{-1} ^{232}Th and similar concentrations are characteristic of its waste [15].

In the ores used at Sillamäe, both the quantities and the activity concentrations of U and Th could be more than tenfold higher than the corresponding exemption levels established in Estonia. On the basis of extensive surveys and assessments, several EU countries (see, e.g., [16]), plan to optimise exemption levels for natural radionuclides used in the non-nuclear industry. The purpose of this initiative is to find an acceptable cost-effective compromise between safety requirements and those of production. A similar process has not yet started in Estonia. Therefore, work activities involving the above raw materials are considered to be of radiological concern, and authorisation by licensing as radiation practice should be started. The radiation protection and safety requirements imposed by the Radiation Act should be fully implemented for these work activities.

3.2. OCCUPATIONAL EXPOSURE

In the production of rare-earths, Nb and Ta, enhanced occupational exposure for workers is considered possible mainly via the external gamma radiation and inhalation pathways [14]. High U and Th concentrations in raw materials may give rise to elevated radon and thoron concentrations as well as to dust enriched in these radionuclides in the workplace air. The results of a survey [24] performed at the Silmet plant show convincingly that both radon and thoron inhaled by workers are of regulatory concern.

The external dose rates at many locations in the production area are about two orders of magnitude higher than those caused by the natural background exposure at Sillamäe town and its surroundings (0.08-0.18 $\mu Sv\ h^{-1}$) [17]. High external dose-rates up to 36 $\mu Sv\ h^{-1}$ have been monitored on the tailings pond [15], while lower values in the range of 5-10 $\mu Sv\ h^{-1}$ have been recorded for the former mining waste area and the abandoned railway terminal. Near the raw material containers we have measured dose rates in excess of 5 $\mu Sv\ h^{-1}$ [17], which may cause an exposure hazard to workers.

In the above cases, a special expert survey to identify the health hazards should be ordered in accordance with the procedures described above. The survey should include monitoring and assessment of annual average effective doses at the workplaces. Based on the results of the expert survey, the Minister of Environment will decide on the attribution of the exposure to occupational or otherwise. If the assessment results demonstrate that the effective doses higher than 6 $mSv\ a^{-1}$ may occur, systematic individual dosimetry should be introduced for these workers. For occupational doses in the range 1-6 $mSv\ a^{-1}$, the need for individual dosimetry and its implementation will be decided by the ERPC.

3.3. RELEASES AND PUBLIC EXPOSURE

3.3.1. *Water Pathways*

Releases of radionuclides from the Sillamäe tailings pond to the ground water and to the Gulf of Finland have been assessed by the international Sillamäe project [13]. The ground and tap water in the area showed extremely low levels of radioactivity. Releases to the Gulf of Finland constitute the major pathway, specifically the consumption of fish. The uranium isotopes ^{234}U and ^{238}U dominated the source term (about 6.2 $GBq\ a^{-1}$ or 0.025 % of the total U per year). The source term for the unmeasured ^{210}Pb, which was considered to be the main possible contributor to the pathway, was assumed to be equal to that of ^{226}Ra (~0.05 $GBq\ a^{-1}$). Our unpublished results on the highly weathered and leached waste samples from the tailings pond indirectly support the last assumption, as well as a higher leaching rate of uranium in comparison to radium and lead.

The compartmental modelling of the marine pathway gives a relatively low radiological impact to the population. The individual and collective committed doses of the order of 1 μSv and 1 manSv, respectively, have been found for both the current low leakage and sudden dam collapse scenarios [18]. The results of the expert assessment confirm that the water pathways of radionuclides from the Sillamäe tailings pond appear to be of no significant regulatory concern.

3.3.2. *Atmospheric Pathway*

The available data for the determination of the source term for the radionuclides released to air by the Sillamäe site are rather scarce. In ground-level air, dust loads in the range of 0.02 to 1.6 mg m^{-3} have been measured in the town air in 1995-1996 (V. Nosov, Silmet (1997) private communication). Aerosol radioactivity concentrations in the range from LLD to 40 mBq m^{-3} with a mean value of 4 mBq m^{-3} were also determined by total beta counting. At Sillamäe, the mean dust load of 0.23 mg m^{-3} appeared to be significantly higher than the corresponding UNSCEAR world-wide mean value of 0.05 mg m^{-3}. At locations near the sea, as a rule, considerably lower dust concentrations were found. The sources (plant, tailings pond, power plant, etc.) of the suspended dust particles, as well as their radionuclide composition, have not yet been identified at Sillamäe. If raw material or waste particles contribute to the composition of dust, the annual individual inhalation doses could reach tens and even hundreds of μSv. In addition, our analysis of the radionuclide concentrations in soil profiles collected at Sillamäe demonstrate enhanced (relative to the bottom layers) ^{226}Ra and ^{238}U content in the surface soil layers. A similar phenomenon near the oil-shale-fired power plants in northeast Estonia has been attributed to long-term fly-ash deposition [23]. For this reason a careful monitoring and the assessment of the radiological impact caused by the observed dust loading should be performed.

Radon progeny released from the tailings pond and inhaled by the population has been considered to be the major radiological hazard at Sillamäe. Outdoor radon concentrations in the ranges of 200-600 Bq m^{-3} and 14-130 Bq m^{-3} were measured on the wall of the pond and in the town, respectively. According to the expert evaluation (without modelling), the annual inhalation doses from radon progeny originating from the tailings pond may reach 200 μSv [15]. Population doses of this magnitude are of regulatory concern and additional monitoring and assessment should be applied [11].

3.3.3. *Modelling of Population Doses Caused by Radon*

In the Institute of Physics, University of Tartu, we have made a preliminary independent attempt to assess the radiological impact of radon progeny released from the tailings pond and its geographical distribution. For this purpose, Gaussian plume modelling software CAP88pc2 (by permission from DOE, USA) approved by the Environmental Protection Agency (EPA) under the authority of the Clean Air Act has been used.

The source term of about 0.5 MBq s^{-1} of ^{222}Rn was evaluated using analysis results from samples collected from the pond walls in 1993 (both our unpublished results and those from reference [15]). The exhalation rate was calculated using our data on the radon emanation factors (about 12 %) for the waste samples. The available long-term meteorological data for Narva (about 20 km E of Sillamäe) were adjusted for the software. Simulations for distances up to 10 km from the tailings pond modelled as a non-momentum area source (dry area of 170 000 m^2)

were performed. The results of the simulation of air concentration and dose rate distribution are shown in Figs. 1 and 2, respectively.

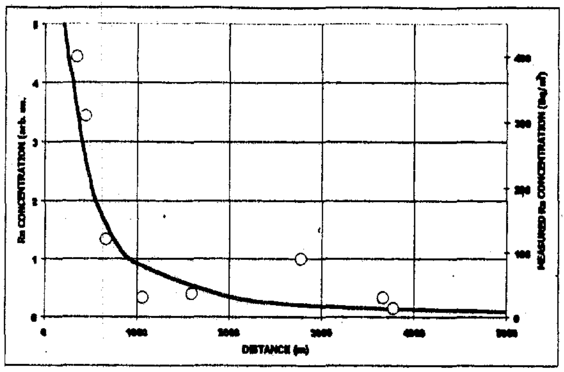

FIGURE 1. Radon concentration in outdoor air vs distance (in the direction ESE) from the Sillamäe tailings pond : solid curve - simulation by using CAP88pc2 and dots - measurements [15]

FIGURE 2. Geographical distribution of annual doses *($\mu Sv\ a^{-1}$)* caused by Rn released from the tailings pond. The isodose curves are the results of simulation (*See text for the details!*).

Figure 1 presents the dependence on distance of the measured radon concentrations in the Sillamäe outdoor air [15] and the corresponding calculated concentrations. A good qualitative correlation of the measured and simulated dependencies is observed, while the simulation is characterised by significantly lower radon concentration values. The reason for the observed discrepancy is not clear and further detailed studies are desirable. An underestimation of the radon source term, a possible influence of the other radon sources (e.g., abandoned mines and/or mining waste area), differences in local geographical and meteorological data (differences in the wind-rose in comparison to Narva), etc., are only a few possibilities.

In Fig. 2, the geographical distribution of the annual doses caused by radon and its progeny in the form of isodose curves superimposed on the map of Sillamäe (*AS Regio, Estonia*) is demonstrated. Maximum annual doses higher than 100 μSv are evaluated near the tailings pond . A rapid decrease in doses is observed as the distance from the source increases. The region with annual doses higher than 10 μSv extends up to a radius of 1-2 km from the tailings pond.

At present no regulatory limit for release rates of specific radionuclides to air or to water has been established in Estonia. However, as follows from the decree [11], the reference levels relevant to the verification of public exposures should help to control the releases. A similar verification procedure for occupational exposure is currently in preparation.

4. Conclusions

The present legal infrastructure for radiation protection in Estonia involves requirements for regulatory control of work activities involving enhanced exposures to natural radiation. In general, the principles, limits, criteria, and methods of regulation are harmonised to the corresponding international ones. More detailed requirements for the regulation of occupational exposure, waste management, etc. are in preparation.

Work activities at Sillamäe involving natural radionuclides cause exposures of concern for both workers and population mainly via the atmospheric pathway and/or external exposure. After careful surveys and evaluations, the authorisation of the work activities as radiation practices is needed to ensure radiation safety at Sillamäe.

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LEGAL RESPONSIBILITY AND LIABILITY FOR THE SILLAMÄE SITE

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1. Introduction

The aim of this paper is to present those international agreements and recommendations which could be applicable to the Sillamäe site, to try to analyse the issue of liability and responsibility in the light of international law in general, to say something about national legislation *de lege ferenda* (how it should be) and to indicate the interaction between legal rules and technical or scientific specifications.

Outside the town of Sillamäe in northeastern Estonia, uranium and other metals are stored in a tailings pond of about 400,000 square meters in area and 25 meters high, some tens of meters from the beach of the Bay of Finland. The residues have their origin in activities mainly carried out during the Soviet Union period for Union purposes, i.e. not for local, regional, or Estonian national purposes. A recently established enterprise, the Silmet Group, is producing rare earths and rare metals at the site. The waste from the new activities, a few hundreds of tons per year, is negligible in comparison with the Soviet Union waste, hereafter called the historical waste, approximately 8 million tons.

2. International Conventions and Recommendations

2.1. THE 1982 CONVENTION ON THE LAW OF THE SEA.

In general, the marine protection articles in this convention pledge the states parties "to protect and preserve the marine environment". Although the terms "pollution" and "dumping" are broadly defined, it is apparent that activities leading to such effects are not totally prohibited under the terms of the treaty, merely sought to be controlled and minimised "to the fullest extent possible." Thus, states parties are directed to take measures, both individually and jointly (on a regional and global basis) to control and minimise all sources of marine pollution and are in effect referred back to *existing marine pollution control treaties,* such as

2.2. THE 1972 LONDON DUMPING CONVENTION.

According to Article IV (III) in this convention, the dumping (i.e. any deliberate disposal at sea of wastes or other matter from vessels, aircraft, platforms or other man-made structures) is *either* prohibited (matter listed in Annex I) *or* requires a prior special permit (matter listed in Annex II) *or* requires a prior general permit (any other matter). A permit shall be issued only after careful consideration of all the factors set forth in Annex III, including prior studies of the characteristics of the dumping site as set forth in the sections B and C in that Annex.

For the Sillamäe site a still more specific international agreement is applicable, namely

2.3. THE 1974 CONVENTION ON THE PROTECTION OF THE MARINE ENVIRONMENT IN THE BALTIC AREA (HELSINKI)

As regards dumping, this convention goes further than the London Convention as it *prohibits all dumping in the Baltic area* except for mud masses and for certain emergency cases (Article 9). According to Article 6 states parties shall take all steps needed to control and carefully limit (mitigate) *land-based pollution* by harmful matters, listed in Annex II. With this aim they shall, inter alia, as appropriate, co-operate in the development and application of special programmes, guidelines, terms and regulations regarding releases, the quality of the environment and products containing such substances and their use. As for *pollution already occurred* (Article II), such as the release of oil and other harmful substances, states parties shall take steps and co-operate according to Annex VI of the convention in order to ward off or as far as possible mitigate such pollution.

2.4. THE VIENNA AND PARIS CONVENTIONS ON THIRD PARTY LIABILITY FOR NUCLEAR DAMAGE

There is no risk of a nuclear accident in the meaning used in these conventions.

2.5. THE 1993 RIO DECLARATION

Principle no. 13 says that states shall develop their national legislation regarding the liability for and compensation to the victims for pollution and other environmental damages. States shall also co-operate promptly and decisively in order to develop and broaden the rules of international law on liability and compensation for environmental damage, caused within the area of their jurisdiction or areas under their control, to be valid *also for such damages in areas outside their responsibility.*

It should be observed that this principle as well as some articles in the Baltic (Helsinki) Convention urge states to act even when the origin of the pollution is outside their primary responsibility.

3. Some Words about EU Law

The Polluter-Pays Principle, the principle that it should be the polluter and nobody else who shall pay for what he directly or indirectly causes, was adopted by the European Community in 1973. The background to this principle is of course that similar rules should be applied in all member states in order not to cause different competitive situations in the countries. Regarding *already existing activities and already existing products,* exceptions may be admitted, provided that the general rules for subventions are followed. One category of subventions allowed could be contributions to *common environmental facilities,* where, within the immediate future, it is not possible to have the expenses completely covered by the polluters, provided that the connected industrial entities will pay their full share in the end. Another exception could be the financing of especially high costs that must be imposed on certain polluters in order to obtain an extraordinarily high grade of cleanup. A third exception could be contributions to stimulate the research on and development of better technology for environmental protection.

The Polluter-Pays Principle also holds in the two directives on waste management, no 75/442 on waste in general and no 87/319 on poisonous and harmful waste. Neither of these directives applies to radioactive waste. There are regulations and directives on shipment of radioactive waste and other substances, protection of workers from radiation exposure, radioactivity in food stuffs etc. under the EURATOM treaty.

As a general remark I cannot see that the EU rules are applicable to the historical waste.

4. Recent Developments in Swedish legislation

The whole system of Environmental Protection legislation has been revised and collected in one major piece of law, the Act (1998:808) on the Environment, which will enter into force in 1999. Chapter 10 of this act regulates inter alia the *Responsibility for investigation and post-treatment of polluted areas, both ground and water areas as well as buildings and installations which have been as polluted as to entail possible damage or other nuisance on the health of people or on the environment.* Responsible for taking such measures shall be the person, legal or natural, who performs or has performed activities that have contributed to the pollution. If no one thus responsible is to be found, then anyone who acquires the real estate in question and knows or ought to know about the pollution (with regard to the obligation for a buyer to examine what he buys according to civil law) will be responsible for the post-treatment. A person who acquires a real estate for his private living will be responsible only if he knew about the pollution. The responsible person or persons shall, as appropriate, take those steps that are necessary to prevent or mitigate damage to the health of people or to the environment. When considering the extent of the responsibility, due regard shall be taken to the period of time passed since the pollution took place and other circumstances. Even if the possessor is not obliged to act, he may still be liable to contribute to the costs for the post-treatment with a sum of money, corresponding to the increase in value which the real estate may obtain on account of the post-treatment.

5. Liability of States

A condition sine qua non is that it must be in the genuine interest of every state that borders the Baltic Sea to prevent or reduce pollution of the sea. In a wider perspective, other states are also concerned. One legal issue to be determined is whether a legal duty is laid upon one or more parties/states and not shared by the others or if the liability should be proportionally shared to take the appropriate precautions.

As far as I know, the Sillamäe site during the period of the Soviet Union was regarded as a Union concern and never as a regional Estonian concern – even less as a concern of any state outside the Union or of an earlier member of the Union. Accordingly, the Union is to be considered responsible for the site. If states members of the Union could be considered to have inherited any obligations from the Union that could hardly be the case for Estonia, which according to the prevailing opinion was occupied by the Soviet Union. While the Russian Federation played a major role in the Union, the other member states formally were equal in status.

As has been mentioned earlier, international conventions and the Rio declaration urge states to co-operate, but no detailed rules are given in those documents or in other sources of international law to make the obligations of states in connection with the Sillamäe site more concrete. Such rules, if needed, would have to be convened in mutual agreements. I doubt that in that work much guidance may be gained in historical comparisons with the Soviet Union. There is, on the other side, one general principle, established during the last decades beside all conventions, namely that a problem, such as the Sillamäe site, should not be left to future generations.

6. National Legislation

6.1. RELEVANT OBJECTIVES

International law regulates relations between states in the international society. Provisions concerning relations between natural or legal persons as well as relations between such parties and the state are laid down in national legislation. These regulations for instance comprise the liability for damage as well as corrective or punishing measures in the case of violation of provisions aimed at protection of the environment or other relevant rules.

To my knowledge, only the republic of Estonia has jurisdiction over the Sillamäe site. Thus, rules on protection against radiation as well as rules to protect workers in other respects, rules on preparedness and health care in case of emergency, rules on compensation for damage, and rules for the restoration of the landscape and other matters must be adopted by the Estonian parliament.

It has been recognised by most states that the responsibility for nuclear and other kinds of radioactive waste in the long run must be assumed by the state where the waste was produced or the state where it is found and not only by any private entity. This principle is expressed in the Estonian radiation protection legislation.

6.2. THE INTERACTION BETWEEN LEGAL RULES AND TECHNICAL SPECIFICATIONS

A legal rule might say that "the best technique available" shall be used or that detrimental effects should be kept to a level "as low as reasonably achievable" (the ALARA-principle), but cannot go further, i.e., to try to define what is meant by that. Further and more detailed rules and/or recommendations, for instance in terms of measurable quantities or qualities, would be of a technical character and should be issued, neither by the parliament, nor by the government or a ministry but rather by a technically competent body, independent from economical interests, which body may also be empowered to judge whether the technical specifications and subsequently the legal prerequisites are fulfilled.

The judgement of such a body should be subject to the possibility of appeal to a court of justice, which should have technical and scientific competence available.

It is indicated in the title of the workshop that some of the waste might be transformed into an asset (to be reused). To the extent this will occur it will no longer be "waste" in a legal sense and the activities with such substances ought to be regarded as mining and/or manufacturing instead of waste management. I am not in a position to present any further views on the interaction between these different activities until plans on the future activities have been disclosed.

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REGULATIONS AND REQUIREMENTS—SWEDISH APPROACH

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1. Introduction

Radiation protection in Sweden has a long tradition. One of the pioneers in radiation protection was Professor Rolf Sievert from Sweden. He was very active in international co-operation and was one of the initiators of international radiation protection standards as expressed by the International Commission on Radiation Protection (ICRP), which was established in the late 1920s. The first radiation protection law in Sweden was enacted in the early 1940s, and since then there has been a smooth adjustment to ICRP recommendations. As a member of the European Union, Sweden is now required to apply its Basic Safety Standards (BSS). This is not a problem, because the BSS are very similar to the ICRP recommendations, which have been used in Sweden for many years.

In the area of radioactive waste, there has been a continuous development of radiation protection principles internationally and in Sweden. This includes many different aspects of protection, and work continues in this area. A summary of the present situation in Sweden is presented here.

2. General Approach to Radiation Protection

In Sweden, the regulatory framework includes laws on radiation protection, nuclear safety, protection of the environment, and protection of workers. The 1988 radiation protection act covers the protection of workers, the public, and then environment from the detrimental effects of radiation.

The competent authorities for radiation protection is the Swedish Radiation Protection Institute (SSI), and for nuclear safety, the Swedish Nuclear Power Inspectorate (SKI). For non-radiological protection, there other authorities, including regional authorities, charged with supervision and developing regulations.

The regulations and guidance given by SSI on radiation protection are based on ICRP recommendations, on the EC's BSS and other Directives, on conventions and on radiation protection acts and ordinances. In general terms, the protection of man and the environment is regulated by requiring justification of a practice, by setting limits on the annual dose to workers (in practice, 20 mSv/year) and to members of the public (0.1

mSv/year), and by requiring the so-called optimisation of protection. Justification of a practice means that the benefits must be larger than the detriments caused by the practice. A part of the detriment can be expressed as the total exposure of humans (workers and public). The collective dose can be estimated per year of practice in units of man-Sv from that year until a given cut-off time in the future. In the case of radioactive waste and its handling, storage and disposal, it is not so much a question of justification. The waste is a part of the overall practice and therefore the doses should be limited and protection optimised. Therefore, in the evaluation of justification of the overall practice, waste management should be included from the beginning.

Sometimes the waste has resulted from an old practice, an accidental or uncontrolled event in the past, etc. In that case, an intervention must be considered that must do more good than harm, i.e., it must be justified. Radiation protection in an intervention should be optimised, and dose limits may or may not apply, depending on the situation.

The justification procedure is applied by the licensee, the authority, or the government, depending on the size of the practice and its importance and significance for society. The justification of nuclear power, for example, is evidently a matter for the government to judge.

Setting limits is a straightforward procedure. Exposure is regulated directly by dosimetric methods and by indirect methods like limits on release rates, on dose rates in air, and on activity concentrations in air and water. Routine checks and control are done by the licensee and are regularly reported to the authority. In the case of abnormal releases, dose rates, or activity concentrations, the authority is informed according to specific rules. In general, actual radiation doses, in direct or indirect terms, are less or much less than the limits. This is particularly the case as regards the exposure of the public.

Optimisation of protection is really the most important part of the regulations on radiation protection. It means constant effort to reduce radiation doses and risks as much as is reasonable, including improvements below the limits that have been set. However, it appears that in real life, strict optimisation is often difficult to achieve and therefore it is not formallly applied. Common sense, good practices and engineering, a sound safety approach, and the necessity of profit in safety and competition are factors that often replace formal optimisation and do so with good results. Formal optimisation is most common in connection with the protection of workers, particularly in the nuclear industry. Protection in each specific case can often not be seen as isolated from the surrounding conditions that directly or indirectly influence the working situation. These may include organisation, supervision, and education, as well as technical factors like good manufacturing and engineering practices and good isolation of sources.

In a formal optimisation using cost-benefit analysis techniques, the detrimental effect of radiation is expressed in terms of collective dose. For occupational exposure, the calculation of collective dose is simple, but in the case of public exposure, this is not always so. Individual and collective doses resulting from long-term releases or leakage into the environment from waste repositories containing long-lived radionuclides are very difficult to assess because of uncertainties in the long-term stability of containment and barriers, of the future environment and the resulting doses to humans. Therefore, complementary methods to judge the level of protection may be applied, for instance, comparison with natural radiation levels and their normal variation or using other safety indicators.

3. Disposal Options in Sweden and Relevant Regulations

Sweden has a number of disposal options for radioactive waste, which are described in this section.

3.1 DIRECT RELEASE

For nuisance wastes from practices using very small amounts of radionuclides or natural radioactive substances and wastes from patients who have been treated with nuclear medicine, direct release is possible without any control or with some indirect control. Often the release is into the municipal sewage system.

3.2 CLEARED WASTE

Cleared waste, that is, waste that is caused by a licensed practice but whose activity is less than a clearance level, can be treated as non-radioactive either for reuse, recycle, or disposal in a municipal refuse dump.

3.3 DIRECT RELEASE UNDER CONTROL AT THE SOURCE

The source is considered to be the release point. Examples are releases from nuclear power plants to the air and water. Samples are taken in the environment to monitor possible contamination. The releases must be as low as reasonably achievable, and resulting doses to critical groups should be less than 0.1 mSv/year, with collective dose less than 5 mSv/year per GW-year installed electrical capacity.

3.4 DISPOSAL IN MUNICIPAL SEWAGE SYSTEMS AND REFUSE DUMPS

This option is possible for relatively small amounts from non-nuclear practices such as hospitals, research institutions, and industry. The radionuclides are mainly short-lived, and disposal is limited to very small amounts per occasion (1ALI), per month (10ALI), and per sealed source (50 kBq) for each individual practice (laboratory, etc.) ALI refers to the minimum value of the radionuclide in question.

3.4 SHALLOW LAND DISPOSAL

Shallow land disposal is allowed for very low-level waste from the operation of nuclear power plants. Such waste disposal facilities are built at the nuclear power plant site and are supervised by the operator. The conditions are, inter alia:

- The total activity in the disposal facility shall be below 100 GBq.
- The specific activity of the waste packages to be disposed at the facility shall be less than 300 kBq/kg of radionuclides with half-life longer than 5 years.

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- The surface dose rate of each package shall be below 0.5 mSv/h.
- No other environmentally hazardous waste is allowed to be placed in the disposal facility.
- The leakage of radionuclides from the disposal facility into the local environment shall not significantly change the existing radiological impact on this area.
- Doses to individuals in a critical group are not allowed to increase above that expected from routine operation of nuclear power plants.
- The total radioactivity in the disposal facility and its radionuclide composition shall be known.
- The total radioactivity in the disposal facility must be so low and must have such composition and half-lives that clearance would be allowed after at most 100 years after closure of the disposal facility, i.e., no control after that time shall be necessary as regards the use of the facility and its environs. In the applicable regulations, there is a requirement for institutional control for 50 years after closure, and the site must be marked on an archived city map.

3.5 INCINERATION

Incineration is allowed for low-level combustible waste from nuclear and non-nuclear facilities. This method implies a reduction of the volume of the waste, which makes the storage and disposal of the residues (ash) cheaper. Incineration requires good filtration of the exhaust gases as well as facilities for appropriate disposal of the ash. A facility for incineration is operated at Studsvik.

3.6 STORAGE AND DISPOSAL IN A REPOSITORY

A rock repository is available below the seabed for low-level and intermediate-level radioactive waste from nuclear power operation and other nuclear applications in Sweden. The facility called SFR-1 is situated close to the Forsmark nuclear power plant. SFR-1 is located about 50 m under the sea bottom and is accessed through two sloping tunnels. Locating the repository under the sea bottom minimises the hydraulic gradient and the ground-water flow into the repository. The more active waste is disposed of in a 50-m-high concrete silo, and less active waste, in rock caverns.

A major condition in the operating license for SFR-1 is that every waste category to be disposed shall be approved by the authorities. Such approval provides good control of the type and amount of waste and the quality of the package. Performance and safety analyses have shown convincingly that future doses to critical groups and the public will be well below the acceptable limits, and the approval of waste categories will strengthen the reliability of the assumptions behind these analyses.

3.7 PLANNED DISPOSAL OPTIONS

An underground rock repository is being planned for decommissioning waste from the future dismantling of Swedish nuclear plants.

In addition, a repository is being planned for disposal of high-level waste and spent nuclear fuel at a great depth (about 500 m) in bedrock somewhere in Sweden. The choice of site and construction of the repository is a complicated procedure of which design work as well as performance and safety analyses constitute one part; confidence building and political and public acceptance constitute another.

The only radiation protection regulations issued for spent fuel and high-level waste include fundamental requirements on:

- Protection of human health. This is expressed as a planned maximum annual risk of 10-6 to representative individuals of a critical group.
- Protection of the environment. This means protection of biological diversity and of sustainable use of biological resources.
- Protection across country boundaries. This means that neighbour countries (or other countries) must not be exposed to higher radiation levels than those accepted in one's own country.
- Protection of future generations. This means that the predictable impact on the health of future generations must not exceed what is acceptable today.
- Burdens on future generations. This means that unreasonable burdens on future generations shall be avoided by good planning of the work, by starting the disposal in a timely way, by restricting possible releases in the future, and by building a funding basis to finance the disposal.

The environmental impact in terms of collective dose shall be assessed quantitatively for possible releases over the first 1,000 years and the resulting doses to people over 10,000 years. Possible releases after 1,000 years and their impact in terms of collective dose must be assessed in a more qualitative way, with different possible scenarios.

Environmental impact assessment (EIA) is a very important part of the overall process of licensing a disposal facility. In addition to performance and safety assessments, it also includes the whole chain of requirements on alternative options, participation of the public in decision making, information, and consideration of all possible influences within the environment. EIA is regulated in ordinances and is very significant in high-level waste disposal.

4. Specific Conditions and Requirements on Mill Tailings

In principle, the same basic requirements apply to mill tailings as to other radioactive waste disposal facilities. Besides the radiological impacts, other non-radioactive toxic substances must be considered, and the impact of these substances may be more serious than that of the radioactive components.

The Swedish experience of uranium mining and milling and of tailings is from the Ranstad open pit plant, which was built in the 1960s. Mining started a few years later and from 1965 to 1969 a total of 215 tons of uranium oxide was extracted from 1.5 Mt alum shale. Development work continued until 1984, when all activities based on alum shale were ended.

The Ranstad restoration is included in a special programme for the decommissioning of facilities and management of waste from earlier government-funded development of nuclear power in Sweden. The programme is financed by a fee charged to the utilities on the basis of their current production of electricity by nuclear power. The Ranstad restoration cost has been estimated at 140 million SEK (about USD 17 million). The distance from Ranstad to the nearest village is 3 km and to the nearest city, 10 km. The tailings cover an area of about 25 ha, and the volume is about 1 million m^3. Building and related activities are prohibited on the remediated area.

Sealing of the tailings has resulted in a major decrease in heavy metal and uranium transport from the tailings, and it is expected that the current clean-up of seepage waters will not be necessary in a few years.

Some special features of the regulatory requirements are the following;

- A problem in remediation planning is the lack of a clear definition of an environmental goal. In general terms, the goal is expressed as a restoration to natural environmental conditions. The requirements for radiation levels are expressed as being within the normal background variation for the area, but the environmental parameters used to decide when and whether the remediation goals are achieved are the radiation levels in the seepage water.
- The methods used to estimate whether proposed remedial plans will meet post-closure goals are deterministic modelling, comparisons with similar sites, scientific and engineering judgements, and tests and studies during operation.
- The collective dose is not assessed as a measure of future detriments or for optimisation purposes.
- The impact of radiation on the ecosystem is assessed qualitatively rather than quantitatively.
- The prescribed environmental radiological monitoring includes air, water, and biota during both pre- and post-closure periods. The duration of monitoring is still to be discussed.

In conclusion, the requirements on the remediation programme for Ranstad can be summarized as follows:

- The leakage of pollutants to the environment should be minimised.
- There should be an adaptation to the natural hydrological conditions.
- Cleanup of water, pumping, and other maintenance work should be unnecessary.
- The remediation measures should be effective over several centuries.
- The tailings area should be adapted to the natural environment, i.e., natural vegetation and open for public use for recreation and sports.

Lessons learned by the authorities are several:

- The effects on local environmental conditions, e.g., the ground water variations resulting from the remediation measures, must be clarified more in the preliminary investigations.
- Quality is essential in the remediation work.
- The long-term function of sealing and other barriers is difficult to predict and must be monitored for a considerable time.
- The goal of quality of the environment is difficult to decide.
- The need for information to the public has been underestimated.

NORDIC EXPERIENCE WITH ENVIRONMENTAL REGULATIONS

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1. Introduction

During 1994-1997 a Nordic project on disposal of radioactive waste was carried out as part of the Nordic Nuclear Safety Research (NKS) program. Experience with environmental regulations discussed in this paper is partly based on the Nordic project and partly on experiences from waste management at Studsvik in Sweden.

2. Disposal of Radioactive Waste in the Nordic Countries

2.1. WASTE DISPOSAL SYSTEMS IN THE NORDIC COUNTRIES

Practice and planning for disposal of radioactive waste differs widely in the Nordic countries (see Table 1). This is mainly because only two of the countries have nuclear power plants, but different approaches to the need for early disposal have also been taken in the various countries [1].

2.1.1. Denmark

Radioactive waste from nuclear research and from other users of radioisotopes are collected, treated and stored at Risø National Laboratory. Spent fuel from the research reactor at Risø is returned to USA.

Most of the stored waste is low- and intermediate-level radioactive waste. Risø plans to store the waste for some 30 to 50 years. Disposal will first take place in connection with the eventual complete decommissioning of the nuclear facilities at the research centre.

Short-lived low- and intermediate-level radioactive waste can be disposed of by relatively uncomplicated methods in near-surface facilities if long-lived α-emitting elements are not present in more than trace quantities. In Denmark the nuclear authorities have not yet specified limits for the content of long-lived α-emitters in waste to be disposed of in a near-surface facility.

It is estimated that the total volume needed for disposal of low and intermediate level radioactive waste in Denmark will be less than 10 000 m^3 including waste from dismantling of the nuclear research facilities.

TABLE 1. Radioactive waste systems in the Nordic countries

Country	Waste origin	Waste treatment and intermediate storage	Waste disposal
Denmark	Research (Risø) Hospitals and other medical facilities	Treatment and intermediate storage of waste at Risø	Preliminary studies on final disposal Possibly repository of l'Aube-type
Finland	Nuclear power plants Research (VTT) Hospitals and other medical facilities	Treatment and intermediate storage of waste at the nuclear power plants	Final disposal of low- and intermediate-level radioactive waste at the nuclear power plants Deep repository for long-lived waste
Iceland	Hospitals and other medical facilities	Users store radiation sources	Hard to find a suitable place for a repository
Norway	Research (IFE in Kjeller and Halden) Oil industry, hospitals, medical facilities	Treatment and intermediate storage of waste at Kjeller	Combined intermediate store and final repository at Himdalen
Sweden	Nuclear power plants Studsvik Hospitals and other medical facilities	Treatment and intermediate storage of waste at nuclear power plants and at Studsvik Wet interim storage of spent fuel in CLAB	SFR at Forsmark Deep repository for long-lived waste

2.1.2. *Finland*

Four reactors are currently in operation in Finland: two pressurized water reactor (PWR) units at Loviisa and two boiling water reactor (BWR) units at Olkiluoto.

The owner of the two reactors at Loviisa, Imatran Voima Oy (IVO), initially made contractual arrangements for complete fuel cycle service from the former Soviet Union, including return of spent fuel. However, at the end of 1994 the Finnish Parliament issued an amendment to the Nuclear Energy Act prohibiting most export and import of nuclear wastes, including spent fuel from nuclear power plants.

The owner of the two reactors at Olkiluoto, Teollisuuden Voima Oy (TVO), has opted for storing and, later on, disposing of its spent fuel in a deep geological repository in Finland. A consequence of the amendment of the Nuclear Energy Act is that IVO has to implement the same principles and time schedule as TVO in the management of spent fuel after 1996, when spent fuel can no longer be returned to Russia. A major part of the preparatory work and implementation is done by a jointly owned company, Posiva.

Conditioning and storage of low- and intermediate-level wastes from reactor operation, as well as wastes from their decommissioning, will take place at the nuclear power plant sites. These wastes will be disposed of in underground repositories in bedrock at the power plant sites.

At present, according to the amended Nuclear Energy Act, the management of all nuclear waste must find a domestic solution. Most of the wastes arise from the operation and decommissioning of the four power reactors in Finland. Limited amounts of radioactive waste arising from research activities as well from hospitals and industry are disposed of in the repository at Olkiluoto.

The designs of the Olkiluoto and Loviisa repositories are somewhat different, mainly because of the local geological conditions. At Olkiluoto the host rock massif favours vertical silo-type caverns, whereas at Loviisa horizontal tunnels are more suitable.

2.1.3. Iceland

No specific radioactive waste disposal plans have so far been developed in Iceland. The country is situated on the Mid-Atlantic Ridge, one of the most active volcanic and seismic regions in the world, and it is anticipated that a satisfactory disposal concept may be difficult to find.

2.1.4. Norway

The Institute for Energy Technology (IFE) operates the only facility for treatment and storage of low- and intermediate-level radioactive waste in Norway; therefore all such waste is collected, treated and stored by IFE. Drums and other containers are stored in a storage building. In addition, approximately 1000 drums of low- and intermediate-level radioactive waste have been deposited in a near-surface repository.

A combined storage and disposal facility for low- and intermediate-level radioactive waste has been constructed at Himdalen. The facility is built in hard rock as a near-surface rock cavern facility, 50 metres below the top of the rock formation, located in a small hill. It is accessible through a tunnel. According to a decision by Parliament, buried plutonium-bearing waste containers shall be placed in storage awaiting a future decision on disposal. The waste that can be disposed of is placed in concrete structures (sarcophaguses).

The Norwegian government has decided that the waste drums in the near-surface repository at IFE Kjeller shall be retrieved and placed in the new storage/repository facility at Himdalen.

The Himdalen facility consists of four rock caverns, one for storage of plutonium-bearing waste and three for disposal of other wastes. The outermost cavern will be used as a storage area. The capacity of each cavern is 2,500 drums, giving a total capacity of 10,000 drums for this facility. In the depository, the waste containers will be embedded in concrete sarcophaguses with water-tight roofs. In the storage area, waste containers will be placed inside concrete bunkers.

The facility will operate up to the year 2030, when it is anticipated that it will be filled to capacity. In the year 2030, a decision will be made whether the storage part containing plutonium-bearing waste should be converted into a repository or this waste should be retrieved. The repository will then be closed but will be under institutional control for a period of 300 - 500 years.

High-level waste in Norway consists of spent reactor fuel from two research reactors. Spent fuel elements are placed in storage pits at IFE Kjeller and IFE Halden. There is still no shortage of storage capacity for spent fuel in Norway, and no decision has been taken on the disposition or other future management of spent reactor fuel.

2.1.5. Sweden

Sweden has 12 operating nuclear reactors.

The Swedish nuclear fuel and waste management company, Svensk kärnbränslehantering AB (SKB) operates systems and facilities for the management and disposal of spent nuclear fuel and other radioactive waste from the Swedish nuclear power plants. A complete system has been planned for the management of radioactive waste from the Swedish nuclear power plants and from Studsvik.

A final repository for radioactive operational waste, SFR, began operation in 1988. It is a repository for low- and intermediate-level radioactive waste, built in the bedrock under the Baltic Sea close to the Forsmark nuclear power plant. A 50-metre layer of rock covers the repository caverns under the sea bed. The first stage of SFR, which is in operation, includes buildings on ground level, tunnels, operating buildings and disposal caverns for 60,000 m^3 of waste. A second stage for approximately 30,000 m^3 is planned to be built and commissioned after the year 2000.

A repository for long-lived waste, SFL, is planned to be situated at about 500 m depth in crystalline bedrock. It will consist of a repository intended for encapsulated spent fuel, SFL 2, and a repository intended for other long-lived waste, SFL 3-5. The two repositories will be built at the same depth, but separated horizontally by about one kilometre. The original plans for SFL also included a repository, SFL 1, intended for spent fuel in glass. SFL 1 will, however, not be built.

SFL 3 will be designed for long-lived low- and intermediate-level waste from Studsvik and operational waste from the intermediate storage for spent fuel, CLAB, and an encapsulation plant.

SFL 4 will be designed for decommissioning waste from CLAB and the encapsulation plant.

SFL 5 will be designed for the disposal of concrete containers containing reactor core components and internal parts.

Strictly speaking, not all the waste forms destined for SFL 3-5 fall into the category of long-lived waste. In fact, only the waste that comes from Studsvik, the core components, and the reactor internals is long-lived. Operational waste and later decommissioning waste from CLAB and the encapsulation plant could in principle be disposed of in SFR. However, SFL 3-5 is intended to receive all low- and intermediate-level waste that arises on the post-closure period of SFR.

2.2. SAFETY ASSESSMENTS

Performance and safety analyses are required in various phases of a project to develop and construct a facility for disposal of radioactive wastes [2]. In the initial phase, general strategic studies aim at determining the major options for the management and disposal of different types of wastes. In that phase the analyses are quite and few data are likely to be available. Similarly the methodology to be relied upon can be quite simple.

In the next phase, disposal and repository options are identified and analysed in more detail to determine their feasibility for a particular purpose. The pertinent facilities and the magnitude of the potential hazards involved determine the role and scope of analyses required. In the case of very low-level wastes or wastes that can be exempted from regulatory control it is not usually necessary to employ sophisticated modelling tools. For other wastes—including low-and intermediate-level wastes and particularly those including significant amount of longer-lived radionuclides—increasingly detailed and concept- and site-specific performance analyses are required later during the repository development project.

In the Nordic countries, the regulatory process calls for a preliminary safety analysis report to be prepared in order to obtain acceptance by the authorities and to receive a permit for construction of the disposal facility. During the construction period, more detailed data are obtained on the characteristics of the waste products, packages, engineered safety barriers, and the site-specific features of the geological host medium. These data are employed in the preparation of the final safety analysis report, which is required for the application to receive a licence to commission and operate the repository.

Furthermore, the extent and type of performance and safety analyses are dependent on the purpose for which they are carried out and on which organisation is conducting the studies. For example, the regulatory body may want an independent performance analysis to evaluate the analyses performed by the facility developer. These independent studies may be less comprehensive and may concentrate on points where additional information is considered necessary, for example, to evaluate the importance of remaining uncertainties and whether these have been adequately covered by the use of conservative assumptions in models and data or by robustness in the facility design.

2.3. ENVIRONMENTAL IMPACT ASSESSMENTS

The system for environmental impact assessment (EIA) in a country depends on legislative structures, the legislated application, administrative practice, and general social objectives. It is therefore natural that the EIA systems differ from country to country, even if the directives of the European Community (document 85/337/EEC) and internationally accepted principles are adopted. For example, there are differences among the objectives for the EIA systems in the Nordic countries [3]: The EIA system in Denmark requires specific assessment of environmental consequences for certain projects. Emphasis is placed on public participation and an open decision process. The EIA systems in Finland, Iceland, and Norway require special assessment of environmental consequences for specified projects. Emphasis is placed on project planning and public participation. The EIA system in Sweden gives the authorities a basis for assessment of the effect on environment, health, safety and general interests in accordance with the Swedish Act on the Management of Natural Resources for a broad spectrum of projects.

There are also differences between the Nordic countries regarding responsibility for the Environmental Impact Statement (EIS). The commissioner of the building project is responsible for the EIS in Finland, Iceland, Norway and Sweden. The regional planning authority is responsible for the EIS in Denmark.

3. Management of Radioactive Waste at Studsvik in Sweden

3.1. INTEGRATED MANAGEMENT SYSTEMS

The waste management systems at Studsvik include facilities for incineration of low-level radioactive waste, scrap melting, treatment of intermediate-level radioactive waste and intermediate storage facilities. Activities in the facilities are managed in integrated management systems that meet requirements on quality, environmental management, and work environmental management. Both authority requirements (from the Swedish Radiation Protection Institute, the Swedish Nuclear Power Inspectorate, the County Administrative Board and the Local Environmental Committee) and voluntary requirements in international standards (ISO 9001 and ISO 14001) are considered in the management systems.

The integrated management systems have the same dispositions as the environmental management standard ISO 14001 and include [4]:

- Quality policy, environmental policy and work environmental policy
- Quality criteria, environmental aspects and work environmental risks
- Legal and other requirements
- Objectives and targets
- Quality program, environmental program and work environmental program
- Structure and responsibilities
- Training, awareness and competence
- Communication
- Documentation
- Document control
- Operational control
- Emergency preparedness and response
- Monitoring and measurement
- Non-conformance and corrective and preventive actions
- Records
- Internal quality audits, environmental system audits and work environmental rounds
- Management reviews

3.2. PROCEDURES TO IDENTIFY CHANGES IN RULES AND REGULATIONS

Studsvik has a radiation protection manager. The radiation protection manager is responsible for identification of applicable changes in radiation protection and safety regulations. A safety manager is, in much the same way, responsible for identification of relevant transport regulations, and an environmental manager is responsible for identifying applicable changes in the non-nuclear environmental regulations.

The non-nuclear environmental documentation is available electronically.

3.3. APPLICATIONS

Permits are usually required from a number of authorities when a new or extended facility is to be built or operations are to begin.

An extension project is at present in progress at Studsvik. The project contains:

- Extension and rebuilding of the melting facility at Studsvik
- Improvement of production logistics
- Improvement of environment and work environment
- New laboratory and test facility in an existing building
- New office areas

The project required permits from the Swedish Radiation Protection Institute, the Swedish Nuclear Power Inspectorate, the County Administrative Board, the Local Environmental Committee and the Local Building Committee. The applications to the authorities included among other things an Environmental Impact Statement.

3.4. EXTERNAL AUDITS AND INSPECTIONS

The company responsible for the waste management at Studsvik, Studsvik Radwaste AB, has certifications under both the quality standard ISO 9001 and the environmental management standard ISO 14001. This means that the certification body (in this case Integria Certifiering AB) audits the integrated management systems twice a year.

However, the supervising authorities (the Swedish Radiation Protection Institute, the Swedish Nuclear Power Inspectorate and the County Administrative Board) also carry out inspections. These inspections may be relaxed in the future for companies with quality and environmental certificates, which will also reduce the inspection charge.

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OUTLINE OF A MULTIATTRIBUTE UTILITY APPROACH TO DEVELOPMENT OF A WASTE MANAGEMENT STRATEGY AT SILLAMÄE

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1. Overview

The chapter briefly discusses a framework for analysis of the waste disposition and management problem at Sillamäe, Estonia. It is a response to the need to develop a strategic waste management plan for the Sillamäe site. A hypothetical objectives hierarchy is presented, along with two possible methods for aggregating scores for designated alternatives. For those unfamiliar with decision analysis as a methodology for problem structuring and solution, this chapter may also be viewed as a brief introduction to some of the ideas behind decision analysis.

Decision analysis is a suitable approach to complex technological choices because the decision and any associated tradeoffs among problem objectives may be modeled as an optimization problem. Optimization has advantages over more heuristic approaches to selection of strategic technological solutions. These advantages include the potential for ordering alternatives by their expected utility scores, sensitivity analysis, and structured revisiting of the problem based on initial (and, if necessary, subsequent) outcomes.

Waste management and disposal problems, particularly nuclear waste disposal problems, have been addressed by many decision analysts [1-4]. The latter citations are examples of Multiatttibute Utility (MAU) Analysis, a decision analysis technique that is most appropriate for evaluation of waste management strategies at Sillamäe.

2. Stakeholders

The list of potential stakeholders with an interest in the process of establishing a site management plan for Sillamäe is long. Some candidates for stakeholder input include (but are not limited to) regional and national Estonian governmental representatives, the SIERG (Sillamäe International Expert Reference Group), representatives from local, regional, national and international interest groups, and citizen groups from areas under consideration for permanent waste disposal.

Stakeholder identification is a critical part of a MAU analysis, and stakeholders should participate in as much of the analysis process as is feasible. Discussion of specific techniques for elicitation of values and objectives from relevant stakeholders [5-6] is beyond the scope of this chapter. While consideration of various conflicting values and objectives is a difficult, potentially contentious process, methods exist for approaching consensus on high-level-objectives [7].

3. Multiattribute Utility Analysis

Multiattribute utility analysis is a technique for systematically evaluating alternative courses of action when situations are characterized by many interested parties, many possible alternatives, uncertain connections between actions and outcomes, and technical complexity. The many financial and non-financial concerns and the potential need for tradeoffs between waste management strategy objectives mean that a MAU analysis is appropriate in this case.

The MAU approach may be generally summarized [4] as

1. Specification of alternatives, objectives and measures
2. Development of the relevant value functions and objective weights
3. Evaluation (ranking) of the alternatives using the performance measures, value functions and weights
4. Sensitivity analysis

The next sections of the chapter are devoted to a brief outline of each of the above steps in the MAU process. Much of the discussion is concerned with the first major step, alternative and objective specification. Examples are given of objectives and aggregation methods for clarification.

4. Waste Management Alternatives for the Sillamäe Site

All waste management alternatives should be generated in the context of *a long-term site management plan that will include disposition of existing wastes and management of future wastes generated by the current and future processing operations at the site.* The key to a remediation and waste management strategy is viewing past and future wastes, along with the recovery of economically viable resources, as part of the same overall problem context.

The Estonian National Government, through its National Environmental Action Plan (NEAP), has stated that "Decommissioning and recultivation of the Sillamäe waste disposal site" is an short and long-term action item. This and other Sillamäe action items are explicitly connected to pollution clean-up and sustainable groundwater policy goals [8]. The NEAP does not specify waste management alternatives for the site, though.

As we show below, NEAP guidelines could be used in the evaluation of alternatives generated via the MAU analysis process. Alternatives should encompass on and off-site processing and disposal of current and future waste materials. For example, it may be quite reasonable to consider an on-site treatment plan for existing wastes in concert with a plan to move wastes generated from ongoing rare-earth processing operations to another site.

There is an inventory of potentially valuable mineral resources within the existing waste depository. The decision to recover resources should be made on its own merits, and any resource recovery activities should occur as part of a waste disposal and management plan. This view of the situation implies that resource recovery will be considered as an independent component of all alternatives [9].

It is imperative that alternatives generated as part of the decision analysis process drive the technological alternatives considered, not vice-versa. Once a set of broad alternatives has been specified, individual technologies that have the potential to meet project objectives may be considered. Narrowly focusing on various technological choices outside of a MAU framework is likely to result in two undesirable outcomes. First, the solution is likely to be sub-optimal. Second, a loss of emphasis on the need for a long-term, strategic waste management plan may occur. Both outcomes are undesirable; the second may be worse than the first.

5. An Example of a Hierarchy of Objectives for Evaluation of Alternatives at Sillamäe

The advantage of an objectives hierarchy is the ability to translate high-level, value driven objectives into related lower-level objectives. Lower-level objectives are connected to individual high-level (fundamental) objectives and are measurable. Fundamental objectives are likely to be broad in scope and may result in many defining lower-level objectives. Actual specification of objectives should occur as a result of an iterative process involving stakeholders and decision-makers. In this section, we present an example objectives hierarchy that is purely hypothetical and is for illustration purposes only.

Policy-grounded objectives reflect fundamental concerns and issues that stakeholders really care about. They may be seen as direct reflections of stakeholder values [7], and, in this case, might include:

1. Health and safety for the area human population
2. Environmental impacts, quality and protection
3. Economic development
4. Technical viability
5. Cost effectiveness
6. Equity
7. Cooperation (between nations and between private and public sectors)
8. Economic value of recoverable resources

The list of objectives above is similar to the objectives for plutonium disposal proposed by the U.S. National Academy of Sciences [10]. That list has been used [3-4] to evaluate options for disposition of both nuclear spent fuel and weapons-grade plutonium in the United States. Following Dyer et. al. [11], we may organize the eight proposed value-driven objectives into three high-level objectives for evaluation of alternatives at Sillamäe:

1. Environmental Health and Safety
2. Operational Effectiveness
3. Cooperation and Economic Development

It should be noted that both high and lower-level objectives should be applicable to the entire range of potential alternatives. A high-level objective such as Environmental Health and Safety should have associated lower-level objectives that may be linked to alternatives that span the range of not moving (or treating) any of the existing and future wastes currently at Sillamäe to moving (and/or treating) all of them to an acceptable site given the lower-level objectives.

Suggested lower-level objectives, which will be associated with specific measures, are listed below in Figure 1. We will also use the term "attribute" to describe a lower-level objective, an objective with an associated scale and measure. In Figure 1, high-level objectives comprise the first two columns, and proposed lower level objectives are under the Measurable Objectives heading.

Fundamental Objectives	Linked High-Level Objectives	Measurable Objectives
Environmental Health and Safety	Human Health and Safety	Radiation levels within x km of site
		Non-radioactive dust exposure levels within y km of site
		Worker exposure to radiation/dust
		Worker accident risk
	Environmental Protection	Transport accident risk
		Aquifer contamination
		Gulf of Finland contamination
		Rare/protected animal protection
		Cultural/archeological sites damaged or destroyed
	Equity	Social equity
		Procedural equity
Operational Effectiveness	Alternative Feasibility	Probability that a technology will be effective
		Total cost (investment/life-cycle)
		Time to project completion
	Value of Recovered Resources	Expected Net Present Value of recovered resources and recovery-specific benefits
Cooperation and Economic Development	Cooperation Between Industry And Government and Between Governments	Level of industry/government cooperation (scale)
		Level of international input and cooperation (scale)
	Economic Development	Number of new, long-term jobs Created
		Development of Estonian Environmental Management Industry

Figure 1. An Example Objectives Hierarchy for the Sillamäe Waste Management Plan

In this example hierarchy, the high-level Cost Effectiveness objective in included in Total Cost as a measurable component of the Operational Effectiveness goal. All other original high-level objectives serve as conceptual links between the three aggregate high-level goals and the lower-level measurable objectives. The matrix used in the Estonian

NEAP [8] to order action alternatives within policy goals is contained in the above objectives hierarchy. Some of the nine criteria used in the NEAP appear as high-level objectives, and some appear as measurable objectives.

Measures, ranges and attribute-specific base cases for each of the lower-level objectives need to be specified. The ranges established for objective measures should be sufficiently comprehensive so that all possible alternatives may be evaluated using the objectives. Some details about each lower-level objective, along with suggested measures for each of the example objectives are given below:

Radiation Levels – Environmental awareness and environmental concerns have been shown to be major issues for the population in Chernobyl area [12]. It is likely that the population in the Sillamäe area is also concerned about radiation exposure. Dose rates for places where people live and work may be the most relevant.

Non-radioactive dust exposure – Dust from the site is a concern, especially in summer. Measure: the standard atmospheric dust concentration measure.

Worker exposure to radiation and dust – The term "worker" refers to individuals employed at the site. This attribute could be decomposed into separate categories if necessary. Measures: the same as above.

Worker accident risk – The expected number of workers killed or injured either per time unit – such as a year – or over the lifetime of the waste disposal and management project is a potential measure for this attribute.

Transport accident risk – A measure for this objective might be the expected number of accidents (possibly of varying degrees of severity) per year (or over the project lifetime) directly associated with the transport of wastes.

Aquifer contamination –The measure should reflect aquifer contamination levels in the area around and including the site that are above established aquifer pollutant limits.

Gulf of Finland contamination – The measure should reflect pollutant concentration in the Gulf of Finland attributable to leaching or spills from the site and surrounding area.

Rare and protected animals – Measure: the number of individual species negatively impacted by the strategic waste disposition and management plan. This may be a source of controversy.

Cultural and archaeological sites – This attribute can be decomposed into protected or recognized sites and sites of general cultural interest. Measure: the number of relevant sites impacted. This may also be a source of controversy.

Equity – Social and Procedural Equity [3]. A Social Equity measure connects the process of alternative selection with the idea that no area or population is subject to excessive health and environmental hazards associated with disposition of the wastes. Procedural Equity is concerned with the degree of public input into the overall process.

Technological effectiveness – Some technological alternatives will have been used in similar situations; some might be untried. Measure: a technological effectiveness score that would have to be developed as part of the overall MAU analysis process.

Total Cost – The Net Present Value (NPV) of all investment and life-cycle costs associated with a waste management strategy. This measure should not include the expected NPV of a decision to recover economic resources.

Time to completion of the project – The projected time to completion for remediation of existing wastes and (concurrently) the projected time to implementation of a waste management plan for ongoing rare earth waste production.

Value of recovered resources – The expected net present value of recovered resources. This objective should be structured so that the decision to recover potentially valuable resources from the waste pit is made independent of any remediation decision. Expected benefits should therefore include jobs and investments that occur as a direct result of the decision to recover economic resources. We assume that resource recovery will not occur unless a waste management alternative is chosen. Other benefits associated with resource recovery, such as pollutant remediation, will be included in other attributes.

Industry/government cooperation – The degree to which government and private industry are able to work together. The measure should be a subjectively assessed scale.

International cooperation – A scale representing the degree to which affected and/or interested governmental entities are able to cooperate in development of a comprehensive alternative for the Sillamäe site should be used as the measure.

Number of long-term jobs created – We suggest a measure such as the number of jobs with tenure longer than one year that are created as a direct result of selection of a long term strategic waste management alternative.

Development of domestic Estonian environmental management industry – This OECD objective [13] could be measured using the percentage of possible outside, private (or semi-private) sector contracts awarded to Estonian firms.

6. Assessing Alternatives Using the Objectives

The process of ranking alternatives consists of the following general steps:

1. Rate each alternative on each attribute/measurable objective
2. Derive the value function for each attribute/measurable objective
3. Determine the preference relations between measurable objectives
4. Use weights to aggregate attributes for each high-level goal
5. Establish the nature of preference relations between fundamental objectives and establish high-level objective weights

6. Compute the overall value score for each alternative

The above list represents a more detailed version of items 2 and 3 in section 3 above.

7. Rating Alternatives on Lower-Level Objectives

A rating for each alternative on each measurable attribute must be compiled. In cases where a probability distribution for an alternative's rating on a given measure is appropriate, distributions should be estimated. Probability distributions are preferred to point estimates when multiple assessments result in multiple ratings for an alternative on an attribute, when there is sufficient uncertainty to make a point estimate for a rating risky or difficult, or both. Historical data may exist to model probability distributions; in some cases subjective probability distributions may be assessed or experts may be used to ensure the accuracy of historical data.

Assessment of subjective probability distributions will normally be done through interactive sessions with appropriate experts. Several established methods for probability assessment exist [14], but it is necessary to use a trained analyst to avoid potential inaccuracies and inconsistencies in expert judgments.

8. Value Functions

Value functions measure the preferences of decision makers regarding the relative importance of different levels of a given attribute over the relevant range (or a sub-range) of the attribute. Value functions may be decreasing, increasing, or constant over the range in question. They may also be linear or nonlinear and are scaled so that the value range for each attribute is the interval between zero and one. The key is the assessment of functions for each attribute that reflect decision-maker preferences regarding the lower-level objective considered.

For example, a decreasing linear function might be appropriate as a model of varying values for expected total costs. A decreasing nonlinear value function might be appropriate for radiation exposure levels. Exposure levels between zero and an above-background standard could be nearly equally (and highly) valued, while exposure levels above an established limit could rapidly decrease in value.

Actual assessment and modeling of value functions is a process that must involve analyst interactions with decision-makers and/or stakeholders. This process, which is built around indifference questions designed to elicit marginal values at different points in attribute ranges, is geared toward an accurate mapping of decision maker preferences over the attribute range in question [15].

Indifference (or trade-off) questions ask decision makers to specify certain values that have the same value as a probability distribution. A decision maker might say that a certain cost of $60 million is equivalent to a distribution where costs of $20 million and $100 million are equally likely. Subject to verification using additional trade-off questions, this would result in a linear value function.

The next step is assessment of preference relations and weights associated with the attributes measured by the value functions. Weights must also be assigned to high-level objectives; such weights represent policy judgments made by policy makers [11] and will be used in the final aggregation and evaluation phase of the process. Policy-maker involvement is critical for implementation of the process and selected alternatives. Decision maker (and/or stakeholder) preference relations among attributes determine the form of the appropriate aggregation method.

9. Aggregation Using Weighted Value Functions

MAU weights are assessed via direct comparisons between scaled measures. Attribute ranges are crucial, as weights generally reflect decision-maker preferences *given the attribute ranges*. Weights are derived from specific answers provided by decision-makers to tradeoff questions between attributes. Assessment of attribute weights therefore involves more and means more than simple relative importance of attributes.

Once weights are assessed, values and weights may be aggregated so that a single value $u(\underline{x}) = u(x_1, x_2, \ldots, x_n)$ for the n-vector of attribute measures for each alternative may be computed. Actual computations of weighted value scores is a two-step process. Each alternative receives a value score for its rating on each attribute; these scores are aggregated so that each alternative has a score for each high-level objective. These scores are then combined across the (three, in this case) high-level objectives to attain the final score for each alternative. We present two aggregation functions below, the additive and multiplicative forms. The choice of function depends on between-attribute preference independence conditions, which also must be assessed [14].

Independence conditions are assessed via additional interactive sessions with decision-makers. A discussion of the ways in which the nature of preference independence between attributes is established may be found in [15]. Note that, regardless of which model is used, for each alternative $0 \le u(\underline{x}) \le 1$ will be true, with the preference for each alternative increasing as its aggregate value score increases.

If somewhat restrictive preference independence conditions are met, the additive utility function may be used to compute the utility score for each alternative [15]. The additive MAU function may be written as

$$u(\underline{x}) = \sum_{i=1}^{n} w_i u_i(x_i) \tag{1}$$

where $w_i, i = 1, \ldots, n$ are the weights and $u_i(x_i)\ \forall\, i$ are the value functions for each attribute level for each of the n attributes. In the case of the additive utility model, weights are constrained such that $\sum_{i=1}^{n} w_i = 1$.

If less restrictive preference independence conditions are met, the multiplicative utility function, of the form

$$1 + ku(\underline{x}) = \prod_{i=1}^{n} [1 + kk_i u_i(x_i)] \tag{2}$$

may be used to aggregate values and weights (denoted as positive scaling constants k_i) [15]. Scaling constants k_i are constrained so that $0 \le k_i \le 1\ \forall\, i$.

To complete the connection between individual attributes and the high-level objectives they define, value scores for the "component" attributes associated with each high level objective are aggregated, and each alternative will have a score for each high-level objective. High-level objective weights are then used to derive the final value score – using the appropriate model – for each alternative. These fundamental objective weights may be interactively elicited from policy-makers in order to maintain their involvement in and acceptance of the analysis process.

10. Sensitivity Analysis

Once an aggregation of weights and values for each alternative has been achieved, sensitivity analysis should be done to assess the impact of key (and, perhaps, all) variables on the final value scores for each alternative. For instance, individual weights in the aggregation model used should be varied and new value scores computed each time a weight is changed.

Point estimates for alternative attribute scores, probability distributions, and/or value functions may be also manipulated to examine the sensitivity of analysis results to these parameters. The key to sensitivity analysis is manipulation of a single variable or input while holding all other variables constant. If the vector of attribute value scores is particularly sensitive to one or more inputs (such as an attribute weight), the input(s) in question may be reevaluated.

Scenario analysis enables the consideration of the sensitivity of alternative orderings to variations in more than one problem variable. In a scenario analysis, attribute weights could be simultaneously varied, with new scores for each alternative computed for each weight and function configuration. Simulation may also be used to analyze the sensitivity of alternative ordering to the attribute weighting scheme [16].

11. Summary

This discussion of the MAU process and its potential application at Sillamäe draws from the framework provided by Keeney and von Winterfeldt [3] and Dyer et. al. [11], [4] in their analysis of the problem of the disposition of nuclear power plant wastes and the weapons grade plutonium supply in the United States. Technical details associated with the outlined steps in the MAU process may be found in Keeney and Raiffa [15].

It is important to approach the issue of development of an overall plan at Sillamäe from the strategic perspective suggested in this paper. If this approach is indeed taken, the analysis (and framework for the analysis) that is an output of the process of alternative selection may be used in other waste management situations in the region. This would advance both the NATO goal of cooperation between governments and the OECD goal of cooperation between governments and industry.

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THE POTENTIAL FOR ECONOMIC RECOVERY OF METALS FROM THE SILLAMÄE SITE

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1. Introduction

The Sillamäe tailings pond contains some 12 million tons of waste material from various industrial activities over the past 50 years. These activities included the production of electricity from local oil shales, the extraction of uranium from local and imported uranium ores, and the extraction of rare earth metals and chlorides from imported loparite ores.[1] The process wastes were initially deposited on an unprepared surface, and later behind a wall constructed of this early deposited material, creating a sandy/muddy mass partially covered by a solution whose pH varies between 6 and 10. This formation is typically called a tailings dam or tailings pond. From the data in Ehdwall *et al.* [3], it appears that the Sillamäe tailings pond, which has a solids volume of 8 million m^3 and covers an area of 330,000 m^2, consists of a lower layer of oil shale ash, a middle layer of uranium tails, and an upper layer of loparite tails mixed with oil shale ash, corresponding to the chronological sequence of industrial activity at Sillamäe. The tailings dam wall consists mainly of uranium tails, but includes some oil shale ash. According to [3], the total solids content of the tailings pond weighs some 12 million tons, although Silmet reports that the pond contains only 4 million tons of uranium tails, 1.5 million tons of oil shale ash, and 140,000 tons of loparite tails. Thus, there is some discrepancy as to the quantity of material within the pond.

Tailings, although of low metal content, can present an attractive source of metals for extraction since the feedstock has already been mined and beneficiated (reduced in size), eliminating the significant costs normally associated with these two activities. However, tailings have the disadvantage that, due to their small particle size, their remaining mineralization may have become chemically altered over time, rendering their metal values unrecoverable. For example, the tails may have oxidized, or the metal values may have gone into solution and become bound up with undesirable

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[1] Loparite ore, $(Na, Ca, Sr, Ce)(Nb, Ta, Ti, Fe^{+3})O_3$, can contain up to 5% (50,000 ppm) rare earth elements [1] or 30 to 36% rare-earth oxides [2]. Hedrick *et. al* [2] report that Sillamäe processed a rare-earth chloride fusion cake produced from loparite ore at the Solikamsk magnesium plant in Solikamsk, Perm Oblast, Russia. African columbite (an iron-niobium-tantalum ore) has also been processed at the site.

contaminants. There is also the possibility that the metal values found in the tailings are there precisely because they are the components of the original ore that are physically or chemically difficult to recover. Thus, while tailings depositories may contain significant metal values, this need not imply that the metal values can be economically extracted.

This chapter provides a preliminary investigation into the potential to economically recover selected metal values from the Sillamäe tailings pond. I begin with a review of the contents of the tailings pond, and then proceed to define economic recovery of metal values and investigate whether such economic recovery is possible from the Sillamäe site. Finally, I list several items for further study.

2. Metal Content of the Sillamäe Tailings

To date there are limited chemical analyses and no mineralogical analyses of the solid material in the Sillamäe tailings pond. Table 1 presents the results of an undated solids sample analyzed by Björn Sundblad of Studsvik AB. The rare earth elements are noted with an asterisk. Earth's crust values are also reported in the table so that the metal concentrations in the tailings can be compared with natural elemental abundance. Note that the rare earths are not that rare: their crustal abundance is greater than that of lead and uranium. Values that are metallurgically recoverable at low concentrations and are of potential economic interest are given in boldface. The sample has relatively high concentrations of arsenic, base metals (copper, lead, zinc), and uranium, indicating that it is probably associated with uranium tails. I have no indication of the representativeness of this sample, nor of its date. Thus, the values contained within Table 1 must be seen as extremely preliminary.

A second chemical analysis was performed by Ehdwall *et al.* [3], and is presented in Table 2. The "Wall" samples are from at least 6 locations along the tailings pond wall. Since the wall is reported to have been constructed from uranium tails, these values also serve as an indication of what might lie within the uranium tailings layer of the pond. The "Old Deposition" sample is presumably taken from material lying outside of the tailings pond wall, deposited before the wall was built (see sampling location W5 in Figure 3.1 of [3]). The "Present Deposition" presumably represents waste produced by Silmet during and immediately prior to 1994, the date of the sample. In Table 2 elements of potential economic interest are again given in boldface.

The three samples in Table 2 reveal that the tailings pond wall and the old deposition are of a similar origin, representing mostly uranium tails, and that the present deposition most likely represents rare earth tails. The older material contains substantially more arsenic, base metals, silver, uranium, and vanadium, and resembles the sample taken by Sundblad (Table 1). The newer material contains more titanium, niobium, scandium, strontium, zirconium, yttrium, and thorium, and a substantial amount of lanthanum. The fact that the different layers of the tailings pond contain differing metal concentrations creates the possibility that the tailings could be selectively "mined" for various metal elements. This lowers processing costs, since

TABLE 1: Elemental Analysis #1, Sillamäe Solid Waste Material (Sundblad)

Element	Value	Standard Error	Earth's Crust
Aluminum (Al)	4.66 %	0.02 %	8.41 %
Calcium (Ca)	4.98 %	0.04 %	5.29 %
Iron (Fe)	2.15 %	0.02 %	7.07 %
Potassium (K)	2.26 %	0.01 %	0.91 %
Magnesium (Mg)	0.729 %	0.004 %	3.2 %
Sodium (Na)	1.06 %	0.01 %	2.3 %
Phosphorus (P)	0.054 %	0.001 %	0.05 %
Titanium (Ti)	0.144 %	0.001 %	0.54 %
Arsenic (As)	248 ppm	6 ppm	1.0 ppm
Barium (Ba)	308 ppm	1 ppm	250 - 629 ppm
Beryllium (Be)	< 2 ppm		1.5 ppm
Cobalt (Co)	**12.2 ppm**	**0.4 ppm**	**18 - 29 ppm**
Chromium (Cr)	**34.8 ppm**	**1.0 ppm**	**112 - 185 ppm**
Copper (Cu)	**122 ppm**	**1 ppm**	**75 ppm**
Lanthanum (La)*	**15.7 ppm**	**0.8 ppm**	**16 - 32 ppm**
Molybdenum (Mo)	< 10 ppm		1.0 ppm
Niobium (Nb)	< 10 ppm		11 ppm
Nickel (Ni)	**23.8 ppm**	**0.3 ppm**	**60 - 105 ppm**
Lead (Pb)	428 ppm	5 ppm	8 - 18 ppm
Scandium (Sc)	**6.05 ppm**	**0.80 ppm**	**5 - 10 ppm**
Tin (Sn)	< 10 ppm		2.5 ppm
Strontium (Sr)	**161 ppm**	**1 ppm**	**260 ppm**
Zinc (Zn)	**467 ppm**	**3 ppm**	**80 ppm**
Zirconium (Zr)	59.5 ppm	0.4 ppm	100 - 162 ppm
Vanadium (V)	**170 ppm**	**1 ppm**	**86 - 230 ppm**
Tungsten (W)	< 10 ppm		1 ppm
Yttrium (Y)*	**11.4 ppm**	**0.1 ppm**	**13 - 30 ppm**
Uranium (U)	87.3 ppm	0.554 ppm	0.91 - 2.4 ppm

Sources: Sundblad, [1], [4], [5], [6]. Rare earth elements are noted with an asterisk. Elements of potential economic interest are highlighted in bold.

TABLE 2: Elemental Analysis #2, Sillamäe Solid Waste Material (Ehdwall *et. al* [3])

Element	Wall	Old Deposition	Present Deposition	Earth's Crust
Aluminum (Al)	2.3 - 5.6 %	5.4 %	0.4 %	8.41 %
Calcium (Ca)	4.2 - 21.9 %	9.6 %	21.2 %	5.29 %
Iron (Fe)	2.2 - 3.6 %	4.0 %	0.4 %	7.07 %
Potassium (K)	1.0 - 2.3 %	2.1 %	0.1 %	0.91 %
Magnesium (Mg)	1.2 - 0.7 %	0.8 %	0.2 %	3.2 %
Sodium (Na)	0.2 - 1.3 %	0.5 %	0.7 %	2.3 %
Phosphorus (P)	0.09 - 0.59 %	0.20 %	0.09 %	—
Titanium (Ti)	0.2 - 0.7 %	0.2 %	4.9 %	0.54%
Manganese (Mn)	0.20 - 0.95 %	0.05 %	0.01 %	0.14 %
Silver (Ag)	**2.0 - 16.1 ppm**	**17.8 ppm**	**< 2 ppm**	**0.08 ppm**
Arsenic (As)	73 - 659 ppm	858 ppm	< 8 ppm	1 ppm
Barium (Ba)	0.02 - 0.27 %	0.03 %	0.03 %	0.025 % - 0.063 %
Beryllium (Be)	< 2.0 - 3.2 ppm	4.9 ppm	< 2 ppm	1.5 ppm
Cobalt (Co)	**3.0 - 37.2 ppm**	**7.6 ppm**	**< 25 ppm**	**18 - 29 ppm**
Chromium (Cr)	**39 - 64 ppm**	**95 ppm**	**28 ppm**	**112 - 185 ppm**
Copper (Cu)	**50 - 323 ppm**	**268 ppm**	**65 ppm**	**75 ppm**
Lanthanum (La)*	**12 - 333 ppm**	**30 ppm**	**10,800 ppm**	**16 - 32 ppm**
Molybdenum (Mo)	<12 - 40 ppm	15 ppm	15 ppm	1.0 ppm
Niobium (Nb)	**22 - 345 ppm**	**58 ppm**	**8,880 ppm**	**11 ppm**
Nickel (Ni)	**30 - 54 ppm**	**42 ppm**	**57 ppm**	**60 - 105 ppm**
Lead (Pb)	76 - 497 ppm	1,050 ppm	279 ppm	8 - 18 ppm
Scandium (Sc)	**3.9 - 7.4 ppm**	**8.6 ppm**	**32.1 ppm**	**5 - 10 ppm**
Tin (Sn)	< 12 ppm	< 12 ppm	37 ppm	2.5 ppm
Strontium (Sr)	**141 - 627 ppm**	**275 ppm**	**7,240 ppm**	**260 ppm**
Zinc (Zn)	**167 - 564 ppm**	**331 ppm**	**46 ppm**	**80 ppm**
Zirconium (Zr)	51 - 153 ppm	111 ppm	579 ppm	100 - 162 ppm
Vanadium (V)	**52 - 221 ppm**	**374 ppm**	**< 25 ppm**	**86 - 230 ppm**
Tungsten (W)	< 12 - 14 ppm	23 ppm	< 12 ppm	1 ppm
Yttrium (Y)*	**8.8 - 23.5 ppm**	**28.1 ppm**	**42.3 ppm**	**13 - 30 ppm**
Thorium (Th)	1.2 - 78.1 ppm	11.3 ppm	3,750 ppm	3.5 - 9.1 ppm
Uranium (U)	35 - 299 ppm	290 ppm	42 ppm	0.91 - 2.4 ppm

Sources: [1], [3], [4], [5], [6]. Rare earth elements are noted with an asterisk. Elements of potential economic interest are highlighted in bold.

there is less dilution of economic metal values with this method of processing than with processing the tails as an agglomeration of old and new deposition.

While we do not have an elemental analysis of the loparite ore processed by Silmet, the firm recovers several of the light lanthanides (La, Ce, Pr, Gd, Pm, Sm, Eu), whose concentrations are not reported in Tables 1 or 2. According to Silmet staff, tantalum in the columbite feed is not recovered, and reports to the tailings at a concentration of 5.8-6.1% Ta_2O_5. There may therefore be other recoverable metals in the tailings pond, and this could be confirmed by additional sampling and a more comprehensive chemical analysis.

3. The Meaning of Economic Recovery of Metal Values

With current technology, it is physically and chemically possible to extract, concentrate, and refine to a usable form most of the metals listed in Tables 1 and 2. However, certain metals are more difficult to separate and purify than others, and in general the cost of extraction rises as metal concentration diminishes. For this reason, metal extraction, or mining, occurs only at certain geological anomalies, where metals have been concentrated to well above their concentration in the earth's crust and where they can be separated easily from the waste material. For example, although the sea contains millions of tons of gold in solution, it is not economic to recover that gold given its extreme dilution. In another example, certain refractory high-grade gold ores in Indonesia remain uneconomic because they cannot be easily separated from the host rock.

Economic recovery, then, invokes the concept that it behooves a commercial venture to spend no more than 100 kroons to obtain an additional 100 kroons in revenues. Gold can be extracted and refined from ores containing 1g/t (1 ppm) gold at a cost of about $10/g, while it can be produced from ores containing 9 g/t gold at a cost of about $6/g.[2] With a current gold price of around $10/g, it is not economic to concentrate gold from ores containing less than 1 g/t, while those of 9 g/t are extremely attractive. For this reason we see low priced metals, such as lead, which has a current market price of $0.01/g, being recovered from ores that contain no less than 4% (40,000 g/t) lead. For metal recovery at Sillamäe, it is not economic to spend more than the market price of a metal or metallic oxide in recovering that metal or metallic oxide from the tailings pond. With the cost of extracting metals from tailings resulting largely from physical handling and chemical processing, the lower the metal grade, the higher the cost of extracting a unit of metal (since more tailings must be treated per unit of metal recovered).

Implicit in the idea of economic recovery is not only the concept that the unit extraction cost should be less than the unit market price of the recovered metal, but that the extraction cost should be far enough below market price to provide a return to the invested capital. This is termed surplus or profit, and all firms must generate a sufficient profit to reward capital owners for their investment. Economic recovery

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[2] Prices and costs are given in 1999 United States Dollars (USD) throughout this chapter unless otherwise noted.

requires that the metal be recovered from the tailings pond at less than its market price, with a sufficient margin to provide a return to capital.

Aside from these private or market considerations, we must also evaluate the extent to which a project, such as processing of the Sillamäe tailings pond, contributes to social welfare at the national or international level. For example, if the project leads to the employment of Estonian labor that would otherwise remain unemployed, the additional wage income can be credited as a national benefit of the project. In addition, if processing the wastes makes them safer, or leads to transfers of foreign technology that would benefit Estonians in the future, these benefits must be included. These are national benefits. International benefits might include reduced pollution for neighboring countries. All these benefits must be credited to any extraction program at Sillamäe. On the other hand, if processing the wastes augments their hazard due to increased dust loadings or radon emissions, this cost must be taken into account and charged to the project. Quantification of these benefits and costs is difficult and can require that the analyst put a "price" on the value of a life saved or on environmental amenities. Nevertheless, economists are continually developing tools to quantify these types of costs and benefits, and crude measures are available.[3] Decision analysis, which can avoid the monetization of these costs and benefits by using various welfare functions and indices, is another promising tool for taking into account these non-market factors [7].

Once all the benefits and costs of a project have been enumerated, the main economic tool for analyzing the private or social economic viability of an extraction program is discounted cash flow (DCF). In DCF, the annual cash flows expected to be generated by the project, including the value of all social benefits and costs, are discounted to the present. The discounting accounts for the time value of money and the riskiness of the venture. If the present value of the cash flows is larger than the cost of installing the processing plant, then the project is seen as economic or viable. In other words, the project pays an adequate return to the invested capital. There may be more productive investments available, but the project, at least, produces a positive net return. On the other hand, when the present value of the cash flows, net of initial investment costs, is negative, the project does not generate a sufficient return to warrant investment.

Since a number of remediation techniques for the Sillamäe wastes do not involve metal recovery, it is important to evaluate the private or social economics of a metal recovery plan on a marginal basis. For example, a plan to cover the tailings pond with a layer of cement or clay generates a full suite of social (non-market) environmental benefits, but no private (market) benefits. Based the decommissioning of uranium tailings ponds in the United States, I estimate that covering of the Sillamäe wastes might cost $12 million.[4] If the benefits, in terms of lives saved and environmental preservation, were greater than $12 million, this remediation plan would be economically viable. A plan to extend the remediation effort to recover metal values would generate additional (market) benefits, in the form of a salable product. In turn,

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[3] Consumer surplus and contingent valuation are two popular measures.

[4] The average cost of tailings reclamation for 19 US sites is $0.97/ton [8].

additional costs are incurred, not only in processing the material, but in society's increased exposure to the radioactive elements within the tailings pond. This second remediation strategy must be evaluated on the basis of the *marginal* benefits that it creates—which are private—and the *marginal* private and social costs that the processing incurs. Again, DCF and/or decision analysis must be used to assess the economics of this additional processing activity.

In sum, the economic recovery of metals has multiple meanings. On one level, economic recovery refers to recovery that covers the private or market costs of recovery plus a return to capital. On a broader level, economic recovery incorporates social costs and benefits created by the project, at either the national or international level. In general, with pollution remediation options, social benefits will tend to exceed private benefits, meaning that a project that is not economic on a private level may be economic on a social level. However, it is still possible that a project that is economically viable is not the best use of scarce investment funds.[5]

4. Economic Recovery of Metal Values from the Sillamäe Site

Due to the relatively low metal concentrations found in tailings dams, few dams are reworked by private interests to economically recover their remaining metal values. That is, the private benefits (metal sales) from processing tailings ponds rarely cover the private extraction costs. Exceptions include certain historic gold mine slimes in South Africa which, due to crude processing techniques at the start of the century, contain significant residual gold. Even so, with gold prices now at historically low levels, these tailings processing operations are being curtailed. In the late 1970s, when uranium prices reached record levels ($100/lb. U_3O_8 in 1997 dollars), uranium was recovered from tailings at two locations in Colorado. However, these operations also became uneconomic as uranium prices fell in the 1980s, and were terminated. I know of no currently economic tailings processing operations.

There are several metal concentrations in the Sillamäe tailings pond, particularly in Ehdwall *et al.*'s samples (Table 2), that may render extraction economic. However, detracting from this are contaminants such as thorium, uranium, and arsenic that could devalue any concentrate produced. Without detailed information about the mineralogy of the tailings pond, the extent to which the metal values can be concentrated (e.g., chloride, oxide, or metal) is unknown. For a "best case" estimate, I assume that certain elements can be fully recovered and concentrated to their metallic form, and calculate the in-situ value of these metals in the tailings pond based on existing metal prices. I assume that there are 12 million tons of material in the tailings pond, of consisting of 6 million tons of wall material that resembles uranium tails, 6 million tons uranium tails mixed with oil shale ash (represented by Sundblad's sample and the "old" deposition

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[5] For example, certain of the industrial pollution control expenditures required by the US Environmental Protection Agency have been estimated to reduce pollution and save lives, but at a cost of over $1 million per life saved. Some have argued that a better return on investment could be achieved by allowing higher levels of pollution and spending the savings from not investing in pollution control on preventive medical care for those potentially affected by the pollutants.

in Ehdwall *et al.*'s sample), and 140,000 tons of loparite tails. I have excluded concentration of the uranium in the tails as this element represents a nuisance rather than an economic value at today's prices.[6] In the absence of liquid markets for many of the metals contained within the tailings, prices are often contract-specific and proprietary, and vary with the purity of the product produced. The prices I use are from various sources, although some are several months old and are subject to fluctuation. Prices for certain metals, particularly scandium, depend on the purity of the product produced. I therefore make calculations with and without scandium credits. No value has been attributed to the oil shale ash other than for its metal content. The ash may have additional value.

Table 3 presents an estimate of the gross value of the recoverable metals in Sundblad's sample, which represents the 6 million tons of uranium tails mixed with oil shale ash. While the total value of the metals contained within the uranium tailings is $83 million, or $14 per ton of tailings processed, it is unlikely that the metals could be extracted and refined for a cost of less than $130 per ton.[7] The scandium content is attractive because of its high unit value, but at these concentrations it is unlikely that it can be recovered economically. If the scandium credit is removed, the tails are worth some $5 per ton, the approximate value of the earth's crust. Thus, based on this analysis, there is little scope for economic recovery of metals from the uranium tails. Table 4 performs the same analysis using Ehdwall *et al.*'s "old" sample. Here, silver, scandium, and vanadium are the most attractive metals, but even so the tails remain uneconomic, yielding at best revenues of $25 per ton processed. Treatment of the dam wall, which has a similar chemical make-up to that of the "old" sample (see Table 2), would yield additional material with a similar unit mineral value of about $25/t.

The metal content of the loparite tails is much more attractive. Table 5 shows these metal values to be $428 per ton with scandium recovery, and $385 per ton without. The lanthanum concentration, over 1%, makes this material similar to low-grade rare-earth ores, with the benefit that it has already been beneficiated. In fact, the lanthanum, scandium, niobium, and strontium make up most of the value of these tails. If these elements could be extracted, refined to the metallic stage, and delivered to market for less than $428/t ($385/t without scandium recovery) economic recovery of these metals may be possible from the loparite portion of the tails. To this private market value we must add the monetary value of the social gains of employment and technology transfer to the region, and subtract costs associated with increased pollution due to processing of the tails. A DCF analysis would then indicate whether such processing, which would probably last about seven years, is economically viable. The

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[6] The Energy Information Administration [8] estimated in 1993 that the recovery of U_3O_8 from in-situ leaching operations would cost $30/lb. In-situ leaching is probably close in cost to the technology required to produce uranium from tails, and so this gives a guideline as to the economics of uranium extraction at Sillamäe. Current U_3O_8 prices are around $10/lb.

[7] This is taken from Tschabrun [9], who estimates that the cost of producing rare earth oxides at $75/ton in 1980 dollars. I have inflated this number by 2.8 percent per year, the nominal rate of inflation of plant and processing costs in mining. The actual cost of processing the Sillamäe waste material to an oxide, chloride, or metal requires a detailed engineering study, or at least proprietary cost data from the current rare earth processors, such as Rhône-Poulenc or Molycorp. There is a strong possibility that any refining will charge a penalty for the arsenic and radioactive elements reporting to a Sillamäe concentrate.

TABLE 3: Economic Values, 6 Million Tons of Sillamäe Uranium Tails (Sundblad)

Metal	Concentration (ppm)	Approximate Unit Value ($/kg)	Total Value ($)
Co	12.2	50	3,660,000
Cr	34.8	12	2,505,600
Cu	122	2	1,464,000
La*	15.7	23	2,166,600
Ni	23.8	4	571,200
Sc	6	1,400	50,400,000
Sr	161	9.92	9,584,652
Zn	467	1.20	3,362,400
V	170	7	7,140,000
Y*	11.4	35	2,394,000
Aggregate Value			83,248,452
Value/t			13.87
Value/t without Sc			5.47

TABLE 4: Economic Values, 6 Million Tons of Sillamäe Uranium Tails [3]

Metal	Concentration (ppm)	Approximate Unit Value ($/kg)	Total Value ($)
Ag	17.8	160.77	17,170,418
Co	7.6	50	2,280,000
Cr	95	12	6,840,000
Cu	268	2	3,216,000
La*	30	23	4,140,000
Nb	58	7	2,436,000
Ni	42	4	1,008,000
Sc	8.6	1,400	72,240,000
Sr	275	9.92	16,371,300
Zn	331	1.20	2,383,200
V	374	7	15,708,000
Y*	28.1	35	5,901,000
Aggregate Value			149,693,918
Value/t			24.95
Value/t without Sc			12.50

TABLE 5: Economic Values, 140,000 Tons of Sillamäe Loparite Tails [3]

Metal	Concentration (ppm)	Approximate Unit Value ($/kg)	Total Value ($)
Ag	0	160.77	0
Co	0	50	0
Cr	28	12	47,040
Cu	65	2	18,200
La*	10,800	23	34,766,000
Nb	8,880	7	8,702,400
Ni	57	4	31,920
Sc	32.1	1,400	6,291,600
Sr	7,240	9.92	10,056,939
Zn	46	1.20	7,728
V	0	7	0
Y*	42.3	35	207,270
Aggregate Value			60,139,097
Value/t			428
Value/t without Sc			385

final analysis will depend on the results of a detailed metallurgical engineering study to determine which elements can be concentrated, the extent to which they can and should be refined, and from this the potential markets for the elements.

5. Items for Further Study

Given the limited amount of information on hand at this time, only a very preliminary analysis of the potential for economic recovery of metal values has been possible. The following items are necessary before the economics can be declared with more certainty.

5.1 MINERALOGY, METALLURGY, AND RECOVERY TECHNOLOGY

The elemental concentrations of the Sillamäe waste material provide no information about the mineralogy and potential for metallurgical recovery of the metals. The tailings pond must be extensively sampled and mapped, just as any new orebody, to determine the location of and type of minerals contained therein, their grade, and the potential for dilution during reclamation. In particular, the precise location of the loparite tails, which represent the most valuable portions of the tailings pond but only

1% of the solid mass, needs to be identified so that they can be selectively recovered and processed.

The above "best case" analysis assumed 100% recovery of mineral values to their metallic state. Typical recoveries from low-grade tails are no higher than 90%, and the recoveries possible from the tailings pond need to be determined via metallurgical testing. This testing could also determine potential recovery methods (magnetic separation [10], ion exchange, solvent extraction), concentrate grades, and the extent to which contaminants can be removed. Negotiations with possible refiners (purchasers of a Silmet concentrate), such as Molycorp and Rhône-Poulenc, should take place to determine the desired attributes of any concentrates that they might purchase and treat.

Another issue is the extent to which the feed to any recovery plant will vary as the tails are mined. Extraction processes require consistent feed material. If the loparite tails are dispersed and mixed with other material, their processing may be rendered impossible due to dilution and feed variability.

5.2 RECOVERY COSTS

Given the outcome of the metallurgical assessment, costs of recovery must be determined, as must the stage to which any extraction and concentration will proceed. Initial feedback from Silmet staff indicates that the current operation is fairly high cost, and that economic recovery of the tailings metal values is unlikely. Given the availability of capacity at downstream refiners such as Molycorp and Rhône-Poulenc, it may also be optimal from a net price (price less cost) analysis to only partially concentrate and refine any metals at Sillamäe.

5.3 SOCIAL BENEFITS AND COSTS

The Silmet plant is located in an economically depressed area of Estonia. The monetary value of any additional social gains to employment and technology transfer to the region, and the costs associated with increased regional pollution due to processing of the tails, must be calculated. At issue here is both valuation methodology and the decision as to whether a regional, national, or international perspective is taken. Multiattribute decision analysis may provide a useful methodology that would complement any economic analysis, and this should be explored [7].

5.4 SCOPE FOR EMPLOYING THE EXISTING SILMET PLANT

Silmet is currently utilizing about 20% of its original plant capacity. The unused portion appears to contain solvent extraction capacity, likely to be useful to a tailings recovery program. However, the recovery technologies required for processing the low-grade tails may or may not exist within the unused plant. In this case, the capital cost of any new plant must be estimated and included in the DCF analysis.

5.5 MARKETS

The market for rare earth metals is dominated by a few producers, mainly in China and the US, and a few refiners/marketers, such as Molycorp and Rhône-Poulenc. Currently, given the extreme competitiveness of the Chinese producers, the markets for Sillamäe's rare earth chloride products are weak. Likewise, the market for niobium is dominated by a single producer, Molycorp, and limited mainly to the use of niobium oxide as a steel alloy. This makes market entry difficult and prices uncertain. The markets for scandium and strontium need to be investigated in a similar manner. Mineral commodity experts at the United States Geological Survey are probably the best source for this type of information. Getting Molycorp and Rhône-Poulenc on board would also be beneficial in identifying potential product prices and markets.

6. Conclusions

Preliminary analysis indicates that there is the potential for economic recovery of lanthanum, scandium, niobium, and strontium metals from a small portion of the Sillamäe tailings pond. There are, however, many economic and technical uncertainties that need to be resolved before a more definitive statement can be made.

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OIL SHALE ASH: ITS CHARACTERIZATION AND POSSIBLE USE IN REMEDIATION AT THE SILLAMÄE SITE

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1. Composition of the Inorganic Part of Kukersite

The inorganic part of Estonian kukersite oil shale consists of carbonates and terrigenous materials. Commercial oil shale contains 65 to 80% of inorganic matter. The major component, calcium carbonate, makes up 57 to 75% of the total inorganic matter content. The terrigenous part of the inorganic matter contains fine-grained quarts (8 to 16%), orthoclase (4 to 8%), mica (6%), marcasite (4%), and hydromica.

The ratio of the carbonate and terrigenous components depends on the location of the oil shale deposit. As the inorganic components differ in hardness and strength, grinding of the kukersite oil shale produces dust with particles of different size and composition. Because the carbonate is harder than the terrigenous component, it produces the coarser fraction of dust. The ratio of the carbonate and terrigenous components (C/T) ranges from 0.7 to 1.9 for the finer of ash (particle sizes up to 30 μm) and from 2.7 to 4.0 for the coarser fraction.

2. Composition and Properties of Oil Shale Ash

Research carried out at the Building Materials Research Laboratory of the Tallinn Technical University has shown that fly ash consists of particles with substantially differing properties. The particles differ in size, density, chemical and mineralogical composition, and binding properties [1, 2].

Air separation is considered the most efficient method for industrial fly ash fractionation, both technically and economically. Investigations have shown that it is the most effective way to separate fly ash industrially into "coarse," "fine," and "finest" fractions (Figure 1, Table 1) [3-7].

The coarse fraction particle size usually ranges from 30 to 150 μm. It is most suitable for manufacturing concrete and silicate products [8]. The fine fraction particle size ranges from 15 to 30 μm and can be used to produce the binder kukermite-cement [6]. The finest fraction particle size is mostly less than 15 μm and is used as an additive

TABLE 1. Chemical and Phase Composition of Industrially Used Fly Ash Particle Size Fractions

Component	Quantity of Components in Fly Ash Fractions (%)		
	Coarse	Fine	Finest
CaO	50-56	30-44	29-35
SiO_2	20-28	30-36	30-35
Al_2O_3	6-8	8-11	10-12
Fe_2O_3	4-6	4-6	4-5
MgO	3-4	2.5-3	2.5-3
K_2O	1.5-2.5	2.5-4	4.5-6.5
Na_2O	0.1-0.2	0.1-0.2	0.1-0.2
CaO_{free}	20-32	12-17	7-12
$CaSO_4$	4-8	8-12	14-19
β 2CaO SiO_2	9-14	9-11	8-10
Glass constituent	32-36	28-33	27-32
Pozzolanic constituent	12-18	24-33	25-35
Hydraulic modulus	1.2-1.9	0.8-1.0	0.6-0.8

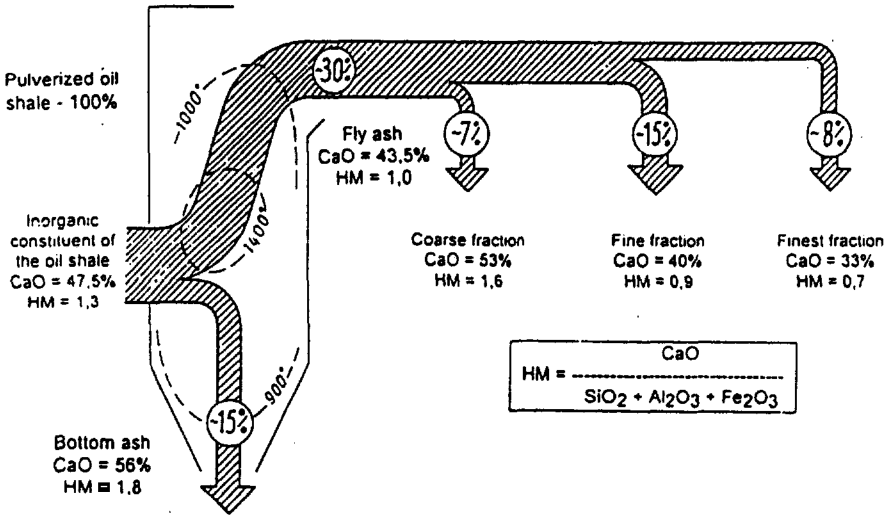

FIGURE 1. Types of Ash Obtained from Boiler Units

in the production of high-grade rapid-hardening Portland-burnt-shale cement [5, 6, 9, 10].

The coarser the fraction, the richer it is in lime (as well as in free lime) and the lower its content of SiO_2, the lower is the content of Al_2O_3. The hydraulic activity of fly ash is determined by the glass phase, the clinker minerals C_2S and CA, free lime, anhydrite, and the pozzolanic constituent.

The physical properties of industrially used fly ash fractions are shown in Table 2.

TABLE 2. Characteristic Physical Parameters of the Fly Ash Fractions

Fraction	Average diameter (μm)	Surface Area (m^2/kg)	Density (g/cm^3)	Bulk Density (kg/dm^3)
Coarse	50-120	50-120	2.82-2.93	1.15-1.25
Fine	12-25	180-250	2.72-2.78	0.90-1.05
Finest	6-9	350-500	2.66-2.70	0.65-0.85

3. Uses of Oil Shale Ash

In the *building materials industry*, oil shale ash has been used in cement and also in gas concrete plants. Oil shale ash gas concrete products have been produced without any cement additive, e. g. wall and ceiling panely up to 6 metres, and wall blocks. However, the concrete should be hardened in an autoclave (similar to gas concrete known as Siporex and Ytong). For the production of this type of concrete, plants were established in Ahtme and Narva, Estonia. In the 1980s, over 300,000 m3 of gas concrete was produced per year in these two plants. In the 1950s, minor quantities of steam-cured cellular concrete were also produced in Tallinn.

In *road building*, fly ash has been used mainly in the construction of sublayers of roads since the 1970s. More than a million tons of ash had been used for this purpose by the end of the 1980s. With the help of autograders, ash was mixed with filler material on the road.

In *agriculture*, fly ash has been widely used for liming acid soils, mainly in southern Estonia. In addition to calcium, ash adds potassium, magnesium, sodium, and a number of microelements to the soil. In the 1970s and 1980s, approximately 70,000 hectares of fields were enriched with ash (~5 tons per ha). To transport the ash and spread it on the fields, pneumatic cement-tank cars were used. Today, because of a lack of financing, this activity is practically nonexistent.

In *environmental activities*, according to data collected by the Department of Environmental Engineering of Tallinn Technical University, oil shale ash has been successfully mixed with purified wastewater. The implementation of this technology has been delayed because of the general unpopularity of adding the wastewater mud to soil, as well as the espense of transportation from Narva to Tallinn, where the largest Estonian wastewater treatment facilities are situated.

4. Activity of the Ash Binders and Burnt Shale Cement

Kukermite is a cement-type binder that has been produced by grinding oil shale ash up to a specific surface of 350 m^2/kg. The activity is measured by the 28-day compressive strength of plastic mortar specimens made of 1 part kukermite and 3 parts standard Volsky sand with flow values from 125 to 135 mm and cured at +20 C.

The kukermite binder hardens very slowly, particularly at temperatures below +10 C. Binder manufactured from the finest fraction is substantially superior in activity to binders manufactured from coarser fractions.

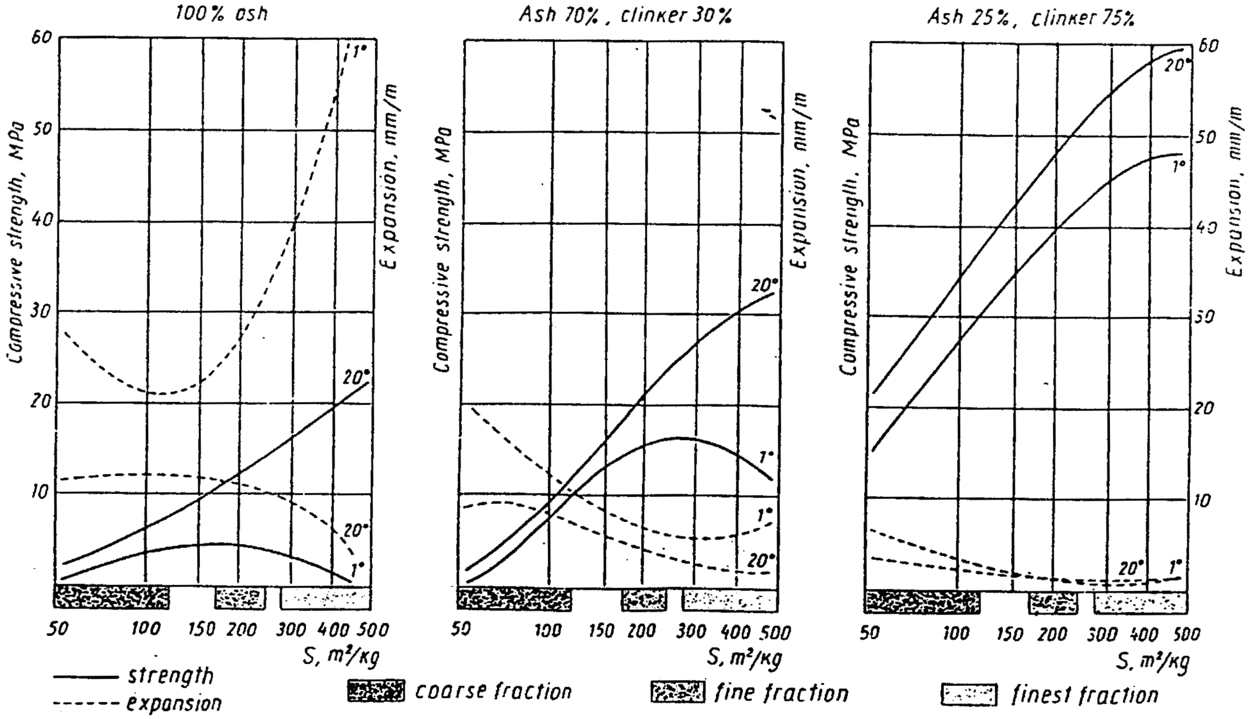

FIGURE 2. Dependence of the Compressive Strength and Expansion of Plastic Mortar of Different Ash Binders on the Specific Surface after 28-Day Hardening at 1 C and 20 C.

The most effective additive that speeds up the hardening of the ash binders, both at normal and lower temperatures, is Portland cement clinker. The efficiency of clinker addition depends greatly on the composition of the fraction used in manufacturing the binder. Basically, the finer the fraction and the greater its activity, the higher is the efficiency of the clinker additive.

The compressive strength and expansion of mortar for different ash binding materials is shown in Figure 2.

Using fly ash as is, the strength and expansion of the final product should be estimated according to the graph on the left in Figure 2. An additional dependence at the hardening temperature 10 C is shown in Figure 3 for the finest fractions of ash. Figure 4 shows the long-term hardening of two compositions of oil shale ash binders: 100% oil shale ash and 70% oil shale ash and 30% clinker.

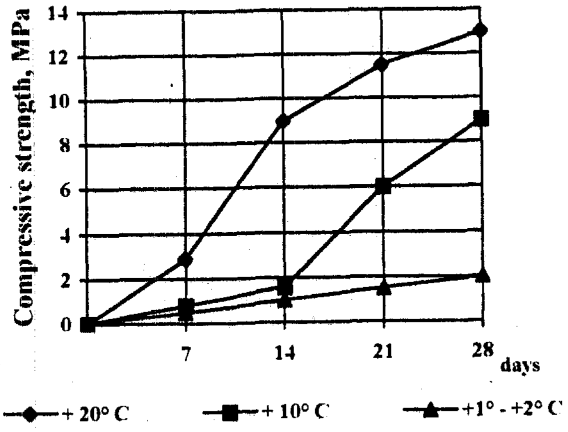

FIGURE 3. Hardening of Oil Shale Ash Binders at Different Temperatures Over 28 Days.

5. Potential for Using Ash Binders in Remediation at the Sillamäe Site

To choose a composition of an ash binder suitable for covering the tailings pond, it is necessary to start from the initial data:

- Minimum compressive strength of concrete for covering,
- Time for concreting,
- Other requirements for concrete: porosity, resistance to water, shrinkage, durability against corrosion, frost resistance, etc.

On the basis of these requirements, the following parameters can be determined:

- Composition of concrete mixture,
- Preparation method for concrete mixture (in a mixer, on an outdoor site),

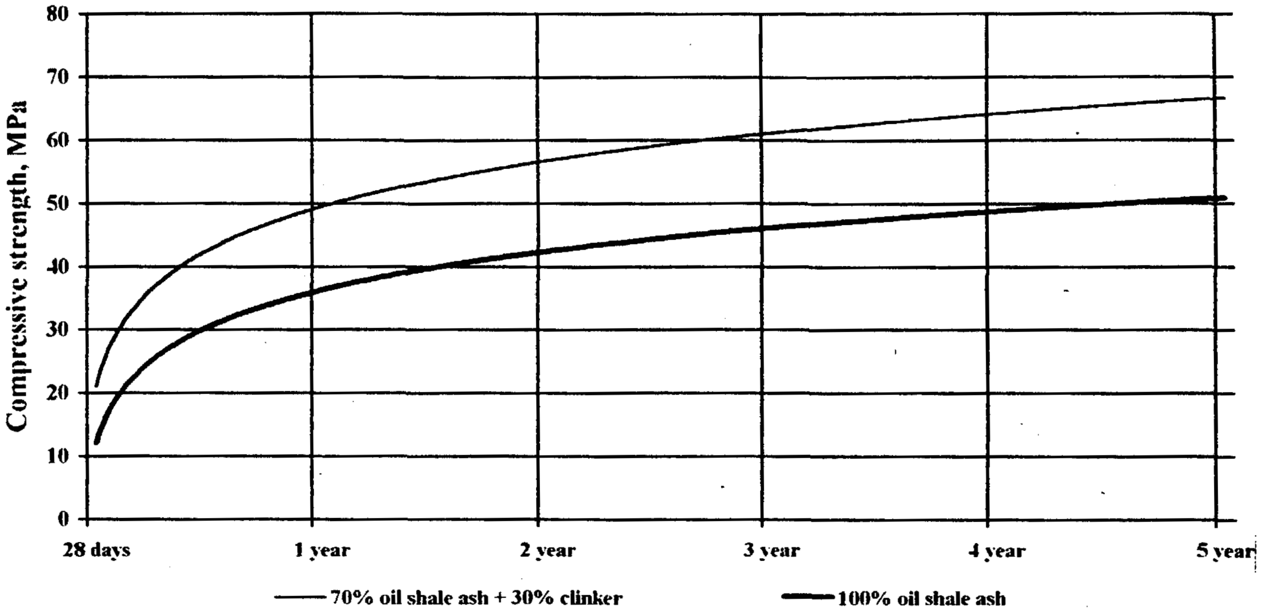

FIGURE 4. Hardening of Oil Shale Ash Binders. Composition 1:3 binder:sand.

- Method of installation (in bulk, mixed with water, single or multilayered),
- Compression (rolling a soil-moist mass, vibration of a more plastic mass.

For the initial data, we propose:

- Minimum compressive strength of the concrete: 15 Mpa (characteristic strength),
- Time for concreting: from April to November (average temperature for April is +2.6 C and November, +5.5 C on the northern coast of Estonia).

An ash binder made of 100% shale ash will not satisfy these requirements due to its characteristic strength (compressive strength after 28-day curing). If in addition to the standard strength of the ash concrete, long-term hardening is taken into consideration, ash concrete could be chosen. For extreme situations (e.g., on slopes or at low air temperature), it is acceptable to use an ash binder manufactured from 70% shale ash and 30% Portland cement clinker. The shale ash must have a specific surface of more than 200 m^2/kg.

It is possible to obtain such ash fractions from power plants in Narva and to produce the binder at Kunda or to transport ash and ground clinker to Sillamäe for mixing the concrete there.

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TYPES OF OIL SHALE ASH AND METHODS FOR INCREASING THEIR REACTIVITY

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1. Introduction

In the Republic of Estonia, electricity is produced by high-temperature (1200-1400°C) combustion of powdered oil shale, which contains 20-25% organic matter and 60-70 % mineral matter (Table 1)[1]. It has a heating value of 8-10 MJ/kg. Because of its high mineral matter content, a large quantity of ash—up to 5-6 million tons per year in all of Estonia—is formed by its combustion [2].

The coarse part of the ash remains in the combustion chamber and is removed as bottom ash, and most of the ash leaves the combustion chamber as fly ash and is separated in several stages (Fig. 1).

Because of the large spectrum of chemical interactions taking place at high temperatures between oil shale particles, different types of ash differ from each other in their chemical and phase composition. These data have been generalized by Kikas et al. [3] and are presented in Fig. 2. This is why differences in the chemical reactivity of the different types of ashes are expected. Chemical reactivity may be an important parameter in selecting the cover materials for remediation of the tailings pond at Sillamäe. The chemical reactivity of the ash can help to prevent migration of contaminants out of the tailings pond.

The objective of the present investigation is the comparative determination of the chemical activity of different ashes formed at the Baltic Power Plant (BPP) and the study of methods for activating them. Because the electrostatic precipitator ash (ESPA in Fig. 1) has the lowest content of free CaO and the highest content of alite and belite as compared to the other types of ash, it has found large-scale use in the cement industry and therefore is less available than the other types. Because of this, only the first three types of ash indicated in Fig. 1 (bottom ash, superheater ash, and cyclone ash) were investigated in this work.

2. Experimental

In addition to the three high-temperature oil shale ash types from the BPP mentioned above, ash from the ash fields (wet ash, WA) and the muds from the hydrotransport water precipitation ponds (PPM) were studied. Activation methods studied were dry (G) or wet grinding (WG), hydration (H), and a combination of dry grinding and hydration

TABLE 1. Average chemical composition of Estonian oil shale components

Organic Matter		Sandy-clay Matter		Carbonate Matter	
Component	**Content %**	**Component**	**Content %**	**Component**	**Content %**
C	77.45	SiO_2	59.2	CaO	53.5
H	9.70	CaO	0.7	MgO	2.0
S	1.76	Al_2O_3	16.3	FeO	0.2
N	0.33	Fe_2O_3	2.8	CO_2	44.3
Cl	0.75	TiO_2	0.7		
O	10.01	MgO	0.4		
		Na_2O	0.8		
		K_2O	6.3		
		FeS_2	0.5		

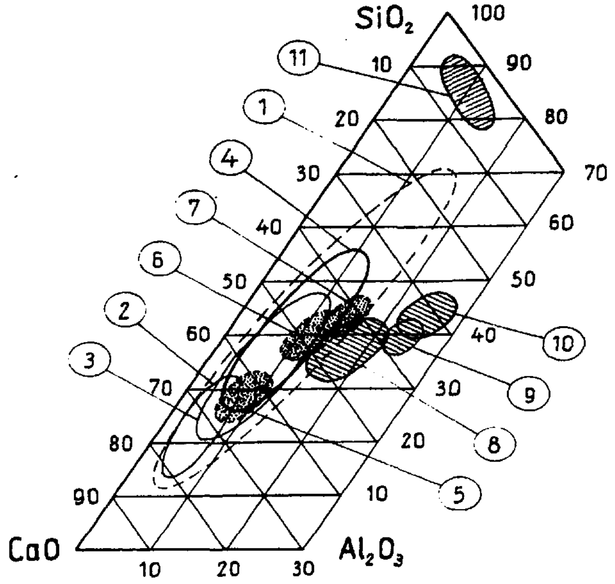

FIGURE 2. Compositions of Estonian oil shale ash, the fly ash fractions and their phases: 1) fly ash fractions selected by density and particle size, 2) bottom ash, 3) cyclone ash, 4) electrostatic precipitator ash, 5) coarse fraction (median diameter 50-120 μm), 6) fine fraction (12-25 μm), 7) finest fraction (6-9 μm), 8) glass constituent of the coarse fraction, 9) glass constituent of the fine fraction, 10) glass constituent of the finest fraction, 11) pozzolanic constituent of all ash fractions.

(G+H) [4]. For comparision, a limestone sample from the Karinu deposit (Estonia) was used.

Different methods of analysis were used to characterise the samples (including chemical analysis, BET nitrogen dynamic desorption, and SEM). Results are presented in Table 2.

TABLE 2. Characteristics of the samples

	Sample				
Parameter	**FA**	**SHA**	**CA**	**WA***	**PPM****
Content, weight %					
CaO(total)	58.6	52.5	49.1	42.0	52.6
CaO(free)	20.1	14.8	13.1	5.5	1.1
MgO	5.8	5.4	5.1	-	0.3
SO_4^{2-}	1.5	7.1	6.1	0.9	3.9
R_2O_3	11.4	11.2	12.6	7.8	1.3
SiO_2 (insoluble r.)	21.7	22.3	23.5	19.8	1.2
Specific surface area, m^2/g					
initial	0.87	0.83	0.62	19.8	4.52
ground	1.45	1.88	1.01	-	-
ground+hydrated	-	4.02	3.33	-	-
Content (wt.%) of the fractional class of - 100μm:					
initial sample	23.7	42.1	58.5	86.0	99.1
ground sample	88.5	90.8	89.8	-	-

* CO_2 - 5.7%, content of CaOfree after calcination at 900°C - 26.5%

** CO_2 - 35.2%, content of CaOfree " - 78.0%

The SO_2 binding rate and capacity were selected as the index for characterizing the chemical reactivity of the oil shale ash. The measurements were carried out in a Q-derivatograph (MOM, Hungary) in isothermal conditions in an air-SO_2 mixture at temperatures of 700-950°C over a period of 30 minutes. A multiplate platinum crucible was used, the weight of the samples was 100 mg each, the thickness of the sample layer 0.2-0.3 mm, the rate of gas flow 270 ml/min, and the SO_2 partial pressure in the dry air-SO_2 mixture 190 mm Hg.

The metal-ion sorption capacity of oil shale ash was determined by using a model system ash-water solution of the cation with mass ratio of solid:liquid of 1:5. The pH of the suspension was 10.0. A solution of Sr^{2+} ion with a concentration of 1000 mg SrO/litre and pH of 6.0 (close to the Sr^{2+} concentration in the waste water of the Sillamäe plant) was used. The suspension was mixed and held at room temperature, hermetically sealed. Changes in the concentration were measured over a period of 7 days by the flame method, using a SP9-7 atomic absorption spectrophotometer (Pye Unicam, UK).

3. Results and discussion

The results of this study, in terms of SO_2 binding rate and capacity of cyclone ash, are presented in Fig. 3. For the initial, untreated, sample of cyclone ash, the SO_2 binding rate in the first seconds of contact at 700°C was 0.20 mg SO_2/mg sample per min (Figure 3A). After two minutes of contact, 9.4 mg SO_2 per 100 mg sample, and after 10 minutes, 13.1 mg SO_2 per 100 mg sample was bound (Fig. 3B). Grinding increased the SO_2 binding capacity by 21% at 2 minutes and 15.3% at 10 minutes.

Previous hydration of the samples was more effective than grinding The effect of water treatment on the sample depends on the contact time between water and the sample. For a time of 0.5 hours, little effect was observed above that of grinding. A 24-hour contact time almost doubled the SO_2 binding rate (0.38 mg SO_2/mg sample per min) over that for untreated cyclone ash, and the SO_2 binding capacity at two minutes by 58% (14.9 mg SO_2 per 100 mg sample). The ash in the Sillamäe tailings pond should display this kind of reactivity because it has been exposed to water for long periods of time.

The best results were obtained using previously ground and hydrated cyclone ash—both the SO_2 binding rate (0.41 mg SO_2/mg sample per min) and the SO_2 binding capacity at two minutes (18.1 mg SO_2 per 100 mg sample) were doubled over that for untreated ash. Wet grinding of cyclone ash enhanced the SO_2 binding rate (0.28 mg SO_2/mg sample per min) and the SO_2 binding capacity at two minutes (13.1 mg SO_2 per 100 mg sample) approximately 1.5 times compared to untreated ash (Fig. 3). Similar tendencies were observed in activation of the samples of superheater ash and bottom ash (Figure 4).

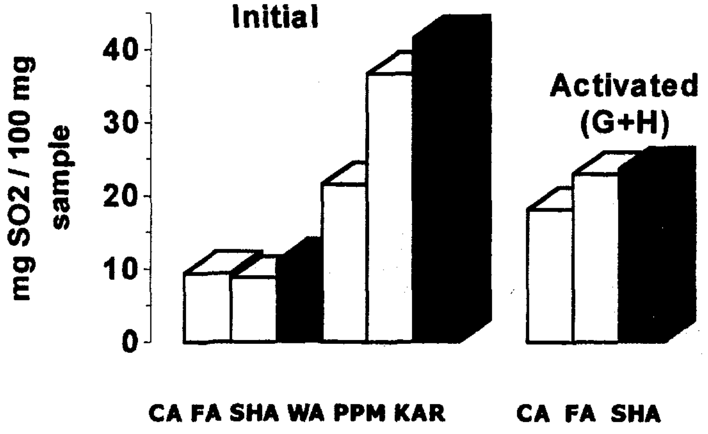

Figure 4. Amounts of bound SO2 (700°C, 2 min) for different samples.

When the temperature of the reaction between ash and SO_2 was increased to 950°C in the first minutes of contact, ash without previous hydration bound less SO_2 than ash that had been treated at 700°C. This change is caused by the decrease in the specific surface area of the samples at temperatures over 900°C. Prolongation of the contact time at 950°C to 2-3 minutes and more led the untreated and the ground samples of the ashes as well as the hydrated ashes to bind more SO_2 than at 700°C (Figure 3).

The most active solid waste from oil shale combustion at BPP, as measured by SO_2 binding, is the mud from the hydrotransport precipitation ponds. X-ray diffraction shows that this solid consists mainly of newly formed Ca and Mg carbonates, with a small amount of impurities, including quartz. The chemical composition of this solid therefore accounts for its high reactivity. Thus, the SO_2 binding rate in the first seconds of contact was 0.74 mg SO_2/mg sample per min, and after 2 minutes of contact, 36.6 mg SO_2 per 100 mg sample was bound. These values are similar to the values for the Karinu limestone (Fig. 4).

All oil shale ash samples showed high reactivity towards Sr^{2+} adsorption. Under our experimental conditions, almost all Sr^{2+} present was bound within a few days (Fig. 5). The adsorption rate depends on many factors and is more rapid for higher solid-liquid ratios in the suspension.

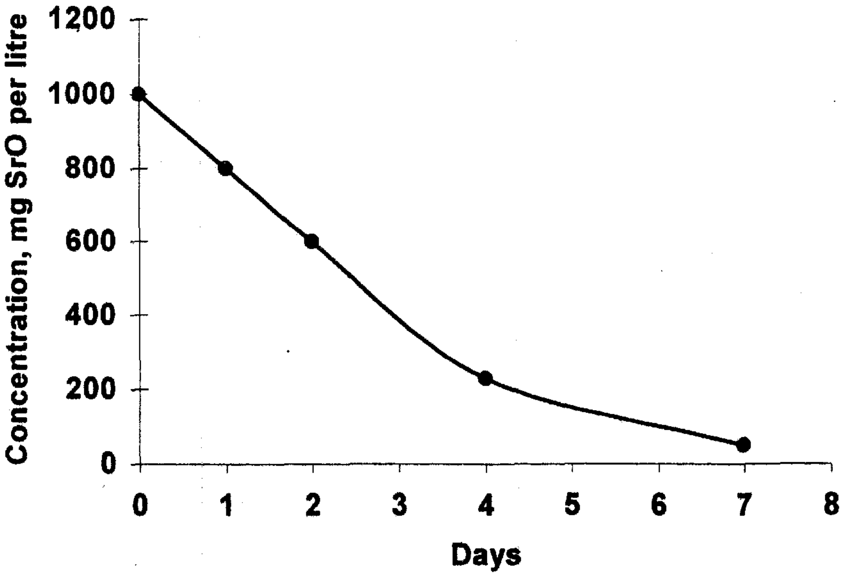

FIGURE 5. Changes in the Sr^{2+} ion concentration.

4. Conclusions

Ash from the combustion of powdered Estonian oil shale in power plants has a high chemical reactivity and ability to bind hazardous components in the waste water from the Sillamäe plant. This characteristic suggests that the ash should be considered as a component of a barrier system for remediation of the tailings pond.

Different types of ash, collected in different parts of a power plant, show different reactivities. Cyclone ash has the lowest reactivity but has the physical properties most suitable for direct use.

Grinding and hydrating ash increase its reactivity significantly. Hydrating takes place during hydraulic transport of the ash as well as during storage of ash under open air conditions. Thus, the ash in the Sillamäe tailings pond should have relatively high reactivity.

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UTILIZATION OF SEMI-COKE FOR ENERGY PRODUCTION

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1. Introduction

Estonian oil shale has been used on an industrial scale since 1916, and for the last 75 years, oil shale has been used for oil production. The organic content of Estonian shale is very high (~40%), one of the world's highest concentrations.

Historically, Estonia has had five different shale oil manufacturing sites that have now been reduced to three: Kohtla-Järve, Kiviõli, and Vaivara. The first two sites belong to Kiviter AS, and the last and newest belongs to Eesti Energia. In addition to these three sites, there is oil manufacturing in Slantsy, Leningrad Oblast, just on the other side of the Narva River, which uses the same feed and technology as Kiviter AS. One of the former oil production sites is the present Silmet plant, where part of the present-day boiler house is the remainder of the oil factory.

The molecular structure of the organic matter (kerogen) in the shale is characterized by a three-dimensional network. Virtually no organic material is extractable from the indigenous shale. Liquid oil products are produced from the shale by pyrolysis in retorts. Industrially, this can be done in several different ways. At present, two different processes are used. A retorting process (Kiviter process) isused by Kiviter, and a rotating kiln process (Galoter process) is used by Eesti Energia in Vaivara.

In the Kiviter process, heated carrier gas from burners placed in the lower part of a vertical retort heats the shale. The shale is fed in the form of the lumps (25-125 mm) at the top of the retort and is pyrolysed while moving down through the retort by gravity. In the Galoter retort, finely pulverized oil shale is heated by recirculated ash heated by the burning of the residual organic content remaining in the shale after pyrolysis. The Galoter retort is a rotating kiln that resembles a Portland cement kiln.

Both processes have advantages and disadvantages. The older Kiviter process has the disadvantage that it can only use lump oil shale, and that the spent shale has a considerable organic content, but its advantage is that the product oil is less contaminated by small mineral particles.

This review describes some problems and possibilities associated with the spent shale from the Kiviter process. Because of the organic content of the spent shale, it is often referred to as semi-coke. This review will also consider problems of utilization of the heaviest oil material rich in solids and water, which is settled from raw shale oil (by-product sludge).

A noteworthy property of Estonian oil shale is its high oxygen content. This leads to a high content of both neutral and acid oxygenated compounds in the product oil. In particular, both mono- and dihydroxyphenols are produced in both retorting processes. Small quantities of these compounds also occur in process water and semi-coke. The phenols are often water soluble or dispersible, and they represent a potential hazard to the environment and human health.

2. Semi-Coke and Other By-Products

The semi-coke at Kiviter AS is a by-product from the Kiviter retorting process in which all liquid and gaseous compounds are evaporated from the shale. During the retorting process, a significant part of the limestone originally present in the shale is thermally decomposed (calcined), forming CO2 and free calcium and magnesium oxides (lime). The semi-coke is currently discharged from the retort via a water seal system.

For each ton of oil shale processed in the Kiviter retort, between 0.55 and 0.60 tons of semi-coke is produced. The semi-coke is disposed in large piles close to the factories by a cableway system. At full production capacity, 1,000,000 tons of semi-coke per year is produced. The Kohtla-Järve and Kiviõli semi-coke deposits contain approximately 69.5 million tons and 12.4 million tons, respectively.

The by-product oil-solid-water sludge is produced in the sedimentation process where small solid particles carried over by the large gas velocities in the retorts are separated from the main raw oil shale oil stream.

3. Properties of Semi-Coke

3.1 MINERALOGICAL PROPERTIES

Since in the retorting process semi-coke is discharged through a water seal, it has an appreciable water content (30-35%). The dry matter of the semi-coke has an organic content of 6-10% and a calcining residue of 78-82%, the balance between the organic content and the residue being CO_2 evolved in the calcination process. The semi-coke contains 1.8-2.2% sulfur. The mineralogical composition of the semi-coke is given in Table 1. Calcium and magnesium are present mainly as carbonates, sulfur as iron sulfide. The semi-coke also contains a number of minor and trace elements, of which the elements with the largest concentrations are given in Table 2.

TABLE 1. Semi-Coke Mineral Content [1]

Component	Concentration (%)
SiO_2	25-30
CaO	30-50
MgO	4-6
Fe_2O_3	6-7
Al_2O_3	6-8
SO_2	5-6
Na_2O	0.1-0.15
K_2O	2-3

TABLE 2. Principal Minor and Trace Elements in Dry Semi-Coke [2]

Element	Concentration (ppm)
As	11
Cl	1600
Cr	30
Cu	70
Mn	440
Rb	50
Ti	1660
V	30
U	3

3.2 ORGANIC CONTENT

The organic content of the semi-coke differs in amount and composition among the different retorts and oil factories. The highest organic content is observed at the new factories (GGJ 5 and GGJ 6), where it is about 10%, while the older factory GGJ 3 and the Kiviõli retorts have an organic content of about 6.5%, measured as percentage of the dry mass of the semi-coke [3].

The organic content in the semi-coke is mostly residual carbon (graphite structures), which are, for example, in GGJ-5 7-8%. Organic substances extractable with solvents are about 0.5% of dry semi-coke.

The water-soluble content of the semi-coke is small, about 0.01% of dry matter. This measurement is from fresh semi-coke. There are indications, however, that some chemical processes proceed in the semi-coke deposits under the influence of temperatures higher than ambient in the presence of oxygen in the air. The appearance of old semi-coke is different from that of fresh material. These processes, however, are not well understood.

The heat of combustion of semi-coke varies, of course, with the organic content and also with the degree of calcination of the mineral component. For semi-coke from GGJ 5, a heat of combustion of 2680 MJ/ton was measured with a degree of calcination of 62%. At a maximum semi-coke production of about 1,000,000 tons/year, such a thermal content would give an average heating power of 86 MW over the year.

3.3 RADIOISOTOPE CONTENT

Despite persistent rumor, the radioactivity of the shale oil is low. The background measured at the Kiviter plant in Kohtla-Järve is 6 to 16 μrad/h, which is very close to normal background radiation. Concentrations of radioisotopes in materials at Kiviter is given in Table 3.

TABLE 3. Radioisotopes in Some Materials at the Kiviter Plant

Material	**^{232}Th**	**^{262}Ra**	**^{40}K**
	(10^{-10}Ci/kg)	(10^{-10}Ci/kg)	(10^{-9}Ci/kg)
Semi-coke	6.8	6.0	21
Sludge	2.1	1.2	1.3
Soil at entrance	4.5	4	7.8

Beta activity, however, varies considerably between sources, as shown in Table 4. The ash is from the power plant at Kohtla-Järve.

TABLE 4. β-Activity in Semi-Coke and Ash

Source	**β-Activity**
	(10^{-9}Ci/kg)
GGJ 5	12
GGJ 4	19.4
GGJ 3	18.8
Ash (Power plant)	26
Fly Ash (Power plant)	43

The activity increases with decreasing organic content, but the increase is larger than would be expected from concentration effects alone. This is especially apparent in comparing the activity of the total ash from the power plant and the fly ash, where the fly ash activity is almost twice that of the total ash. This can be explained only by compositional fractionation that takes place during thermal treatment of the material.

The background radiation levels at the Kiviter plant are probably more affected by the oil-shale-fired power stations in Narva than by the oil processing at Kiviter.

4. Environmental Impact

4.1 WATER

Storage of huge quantities of semi-coke, leakage during oil production, and by-products from oil manufacturing have led to significant pollution of surface and ground water.

Water streams from the semi-coke storage piles contain on average 1 ppm of oil products and 98 ppm of dissolved phenolic compounds. This has led to the presence of oil as well as phenolic compounds in the ground water in concentrations well above the maximum allowable levels. The maximum oil product concentration was measured at 4.6 ppm and phenol concentration at 62 ppm [4].

At present, most of the water streams from the semi-coke storage piles leave the production area without any treatment. Over many years, this had led to the Purtse River becoming one of the most polluted rivers in Estonia.

4.2 ATMOSPHERIC RELEASES

The most important constituents of atmospheric emissions are light hydrocarbons that are part of the sludge generated during production and volatile phenolic compounds originating from the recovery and processing of phenolic compounds.

Atmospheric emissions from the semi-coke storage piles is estimated to total over 1000 ton/year, of whic 250 ton/year is light hydrocarbons, 280 ton/year is oil products, and 43 ton/year is phenolic compounds [5].

5. Utilization of Semi-Coke and Other By-Products

The environmental situations at the shale oil plants are serious. A substantial improvement has to be made before the operation can be recognized as a viable industrial operation meeting minimum European standards.

A large part of the environmental impact of the oil operations appears to be due to the deposition of the semi-coke and other by-products from the oil operation. An improvement in the semi-coke handling process is of vital importance in the turnaround of the oil production process. A strong driving force toward environmental improvement is increased energy efficiency. Today more than 25% of the energy content of the oil shale mined is wasted during production of shale oil.

The energy content of the semi-coke is relatively high but is offset by the high water content in the semi-coke discharged from the reactor through a water seal. Furthermore, energy utilization of the semi-coke is made difficult by the endothermic calcination reaction.

Kiviter AS in cooperation with the Finnish power company Imatraan Voima OY is doing a project study on power generation from semi-coke. The main alternative is to burn the semi-coke in a high-pressure fluidized bed. The benefits of this technology are that it allows the burning of a wide variety of fuels and that the calcination reaction is retarded by the elevated pressure.

Final results from this study are in preparation. Preliminary investigations show that power generating capacity would be between 100 and 200 MW, which would make Kiviter a major player on the Estonian power market. A necessary condition for this development, however, is that the government allow free competition on equal terms between the various power producers.

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MAGNETIC SEPARATION FOR RARE EARTH OXIDE RECOVERY AT SILLAMÄE, ESTONIA

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1. Introduction

The tailings pond at Sillamäe, Estonia, contains waste materials from the chemical processing of uranium ore, alum shale and loparite mining that occurred at the Sillamäe Metallurgy plant through the course of 50 years. The pond contains ~8 million m^3 of wastes and is located on the shores of the Gulf of Finland. The current mineral processing operation focuses on loparite and columbite ores for the production of rare earth oxides [1].

The recovery of dilute quantities of valuable material from mine tailings can be both economically and environmentally desirable. Potential treatments include both chemical and physical processing of the material derived from the mining industry. Magnetic separation is a physical separation process that exploits differences in the magnetic properties of materials [2]. The technology is capable of extracting and concentrating the paramagnetic and ferromagnetic components from solid and liquid streams with minimal pretreatment and at significant rates. In fact, processing rates of 60 tons/h have been achieved in kaolin decolorization. Because this technology is based on a physical separation process in which no additional waste is generated, even complete failure of the separation process results in no degradation from the initial condition; therefore, environmental risk is minimized.

Many rare earth oxides are highly magnetic and can be easily recovered with a magnetic separation process (Table 1). Additionally, because all actinide compounds are paramagnetic, magnetic separation of uranium compounds is feasible. The advent of reliable superconducting magnets also makes magnetic separation of weakly paramagnetic particles attractive.

TABLE 1. Volume Magnetic Susceptibility of Selected Compounds and Elements.

Compound / Element	Volume Susceptibility x 10^6 (SI)
Dy_2O_3	23575.0
Gd_2O_3	13660.0
Eu_2O_3	2676.0
Sm_2O_3	598.0
Pr_2O_3	2422.0
Fe_2O_3	1478.0
UO_2	1204.0
V_2O_3	806.0
NiO	740.0
Sc	263.2
CuO	242.0
Nb	226.0
WO_2	40.2
Th	41.0
UO_3	41.0
Y_2O_3	12.4
CaO	-1.0
Nb_2O_5	-2.0
ThO_2	-7.5
MgO	-11.0
SiO_2	-14.0
Al_2O_3	-18.0
La_2O_3	-19.5

2. Magnetic Separation Methods

Although numerous magnetic separation methods exist [2,3], we have concentrated on development of two distinct methods: magnetic roll separation (MRS) and high gradient magnetic separation (HGMS). These two methods are complementary in the materials that can be treated. Analytical models describing both magnetic separation processes have been developed. The models can provide guidance towards optimization of testing parameters, which is crucial for cost effective operations.

In magnetic roll separation, the most effective separations occur with a dry powder of particle sizes greater than about 75 microns. The separator consists of a thin belt that passes over two rollers where one of the rollers contains a specific arrangement of neodymium-iron-boron rare earth permanent magnets (Figure 1). As the dry powder is fed onto the belt, the paramagnetic particles stick to the belt in the vicinity of the magnetic roller and are deflected. The diamagnetic and weakly paramagnetic particles pass over the magnetic roller with their trajectory relatively unaffected by the magnetic field. This process provides a means for removing paramagnetic particles from the feed

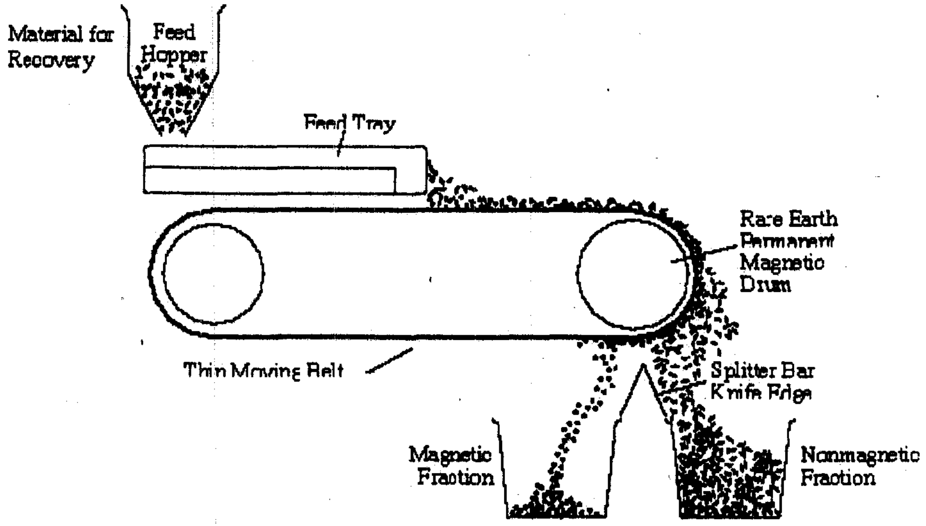

FIGURE 1. Diagram of a Magnetic Roll Separator.

stream. Because MRS is based on permanent magnet technology, it is somewhat easier and less expensive to implement than HGMS. We have investigated MRS for the separation of rare earth oxides (Pr_2O_3, Gd_2O_3), plutonium or uranium from materials such as sand, slag, and crucible fragments (SS&C), garnet, graphite, and gravel.

The HGMS method can be used to separate small particulate magnetic components, down to about 0.2 microns, from solids, liquids, or gases. Most contaminated material is slurried with water and passed through a magnetized volume. Field gradients are produced in the magnetized volume by a ferromagnetic matrix material such as steel wool or nickel foam. Ferromagnetic and paramagnetic particles are extracted from the slurry by the ferromagnetic matrix while the diamagnetic fraction passes through the magnetized volume. The magnetic fraction is later flushed from the matrix when the magnetic field is reduced to zero. HGMS is being developed for the extraction and concentration of paramagnetic particulates (Pr_2O_3, UO_2, PuO_2) from 50 microns down to submicron sizes. For the work presented here, a magnet system is being designed for particulate retrieval from actinide contaminated soils and for the purification of mine-tailing water containing heavy metals. Separator parameters that are significant and can be controlled and optimized for a specific situation include matrix material, applied magnetic field strength, flow velocity, solids density, and surfactant.

3. Magnetic Roll Separation Results

The rare-earth magnetic roll separator is a relatively simple and inexpensive device that can have a processing rate as high as 200 g/min. A systematic approach to address a variety of materials was undertaken, which included laboratory-scale feasibility, optimization of separation variables, and development of an analytical model to predict separator performance.

3.1 MRS MODEL

As we progressed in doing tests on various materials and conditions, it became apparent that there were many different parameters that could be changed and to test all permutations could be unpractical. Therefore a computer model was developed that would assist in narrowing down the relevant and useful parameter ranges for testing.

A magnetic roll separator relies on the relative strength of competing magnetic, inertial and gravitational forces. The force diagram for the model is shown in Figure 2.

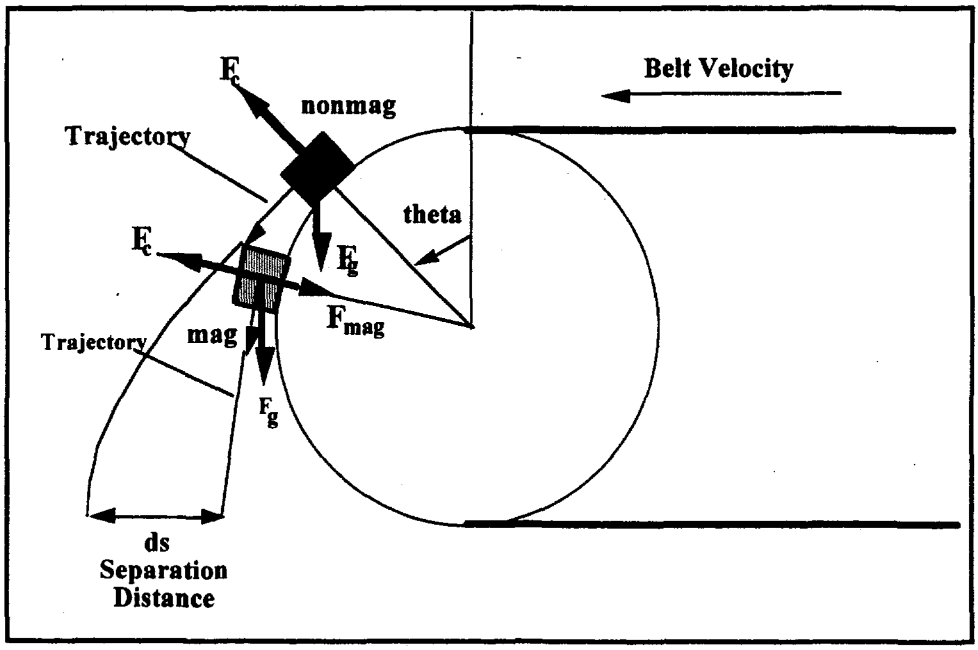

FIGURE 2. Schematic of the Roll Separator Model showing relevant forces and particle trajectories.

These forces are given explicitly for the magnetic, inertial and gravitational terms (electrostatic components are ignored) respectively by:

$F_{mag} = \chi_s B\, dB/dx\, V_p/\mu_0$, where χ_s is the volume magnetic susceptibility of the material, B the magnetic field, dB/dx the magnetic field gradient, V_p the volume of the particle and $/\mu_0$ the permeability of free space;

$F_{int} = \rho V_p\, (D_{roll} + D_p)/2\ \omega^2$ where ρ is the density of the particle, D_{roll} the magnetic roller diameter, D_p the particle diameter; and

$F_{grv} = \rho V_p g$ where g is the gravitational acceleration.

For the particle to stay on the belt, the force (perpendicular to the belt) balance equation is

$$F_{int} = F_{grv} \cos(\Theta) + F_{mag}.$$

When F_{int} exceeds this sum, the particle leaves the belt with a tangential trajectory. In short, a particle that is magnetic will tend to stay on the belt longer than if it was nonmagnetic (everything else being the same) thus giving it a different trajectory.

The model was run with several different ranges of the various parameters. Figure 3 shows the stream separation distance vs. particle diameter for various roller rpm speeds, as an example of the model output. The model predicts that a larger stream separation is obtained with higher belt speeds. From these results, an optimized separation is predicted to occur at higher belt speeds. For each curve, a peak is predicted where the separation can be optimized. Thus, even at slow belt speeds (50 rpm), where inertial forces are small, an optimized separation can occur with a discrete particle range of feed material.

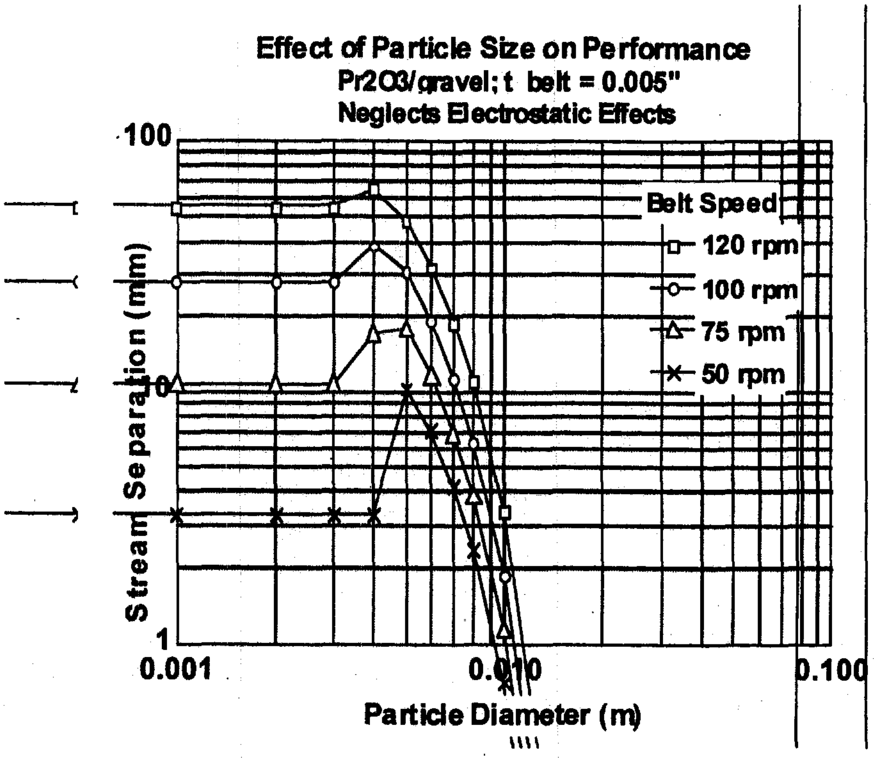

FIGURE 3. Model results showing stream separation as a function of particle diameter and belt speed expressed as roller rpm.

3.2 MRS EXPERIMENTAL DATA

Roll separator experiments were performed on a variety of materials, with variation of belt speed, belt thickness, magnetic roller, material feed rate, particle diameter and moisture content. The goals are to understand which parameters give an optimized separation. The results of one set of experiments, for a mixture of Pr_2O_3 and gravel, are shown in Figure 4, where the variables include both the position of the front splitter setting and the belt speed. The left and right ordinates show the carryover mass for each of the components. Perfect separation would have zero carryover in each component. The 'front splitter setting' is the location of the partition between the catch trays. The data show that a negative front splitter setting allows the separation to be independent of belt speed. At positive splitter settings, the belt speed determines the degree of separation requiring higher speeds to propel the nonmagnetic fraction into the front tray and away from the magnetic fraction, which reports to the rear trays. These results illustrate the importance of separator configuration compared with operational adjustments. Proper separator geometry has a significant influence on performance.

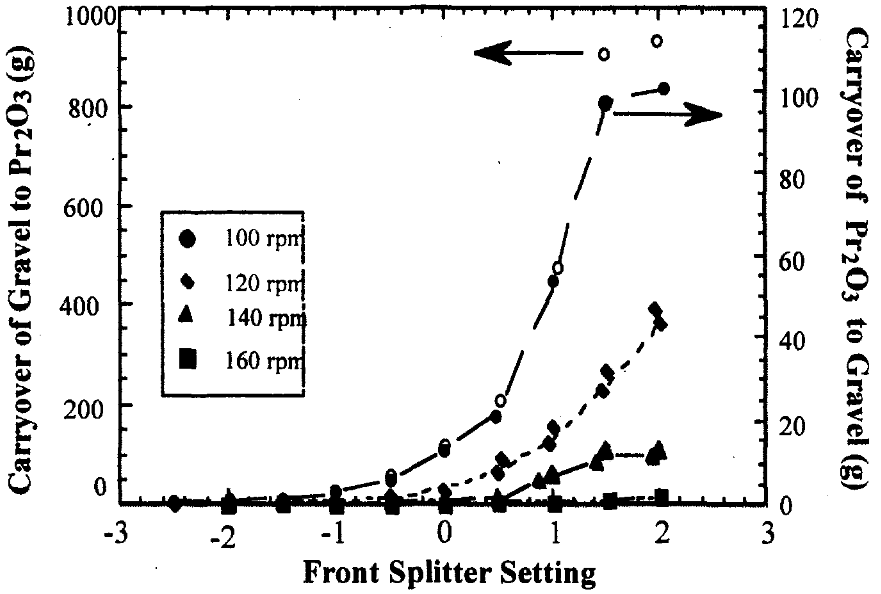

FIGURE 4. Experimental results showing separation as a function of separator geometry and belt speed expressed as roller rpm. Front splitter setting determines the first and second tray partition relative to the magnetic roller.

Table 2 summarizes the MRS results for the magnetic fraction from several types of materials. The table includes percent concentrations of the stream that contains the recovered oxide and the concentration ratio (CR). The concentration ratio is a measure of separator performance. The higher the concentration ratio, the better the separation

TABLE 2. Dry powder magnetic separation application tests summary for the collected magnetic fraction.

Matrix	**Recovered Oxide**	**Particle Size Range**	**% Oxide**	***In* Total Bulk %**	**CR**^{a}
Gravel	Pr	2.5 - 4.25 mm	100	9	10.4
Garnet	U^b	2.5 - 4.25 mm	100	9.4	10^c
SS&C	Pu	106 - 841 □m	76	37	1.8
Graphite	Pu	106 - 841 □m	85	4	4.4

a) CR, the Concentration Ratio, is an indication of the performance. The CR is defined as the ratio of the recovered oxide concentration in the magnetic stream divided by recovered oxide concentration in the initial feed stream. b) uranium metal turnings, c) In the case of uranium separation from garnet, garnet is highly magnetic and the optimized separation occurred at fast belt speeds, which caused the dense uranium metal to land in the non-magnetic tray and the highly magnetic garnet to report to the magnetic tray. Thus, CR in this case is defined as the ratio of the recovered oxide concentration in the *non*-magnetic stream divided by recovered oxide concentration in the initial feed stream.

between the materials and hence the volume reduction. In general, values exceeding 1.5 to 1.7 are of practical importance. The best results on a magnetic roll separator were obtained with residues in which magnetic particles had a high degree of liberation from the host material and the host material was sized before processing to greater than 75 microns. For example, large particulate gravel containing a rare earth oxide (Pr_2O_3) showed, under optimized testing conditions, that a 100% separation is achievable. These results were used to validate and develop the analytical model as described above. Plutonium residues with strongly diamagnetic properties, such as graphite, were also shown to be highly separable; 85% of the plutonium was collected in 4% of the bulk mass. For residues in which the discrete particles were composed of paramagnetic and diamagnetic components (e.g. SS&C residues in which plutonium was included within the diamagnetic slag), a separation was more difficult.

4. HGMS Results

We have completed a comprehensive series of HGMS experiments with radioactive materials. Our results have been used to develop a model that describes the HGMS process [4]. The principal forces governing particle behavior are magnetic, viscous, gravitational and electrostatic. The performance of the high gradient magnetic separator is modeled using a force balance on an individual paramagnetic particle in the immediate vicinity of a magnetic capture site.

In studies involving soil remediation, we worked with Lockheed Environmental Systems and Technology to develop HGMS as part of an integrated system with soil washing and gravity separation equipment [5,6]. The goal is to replace the expensive chemical leach circuit with magnetic separation for treatment of the fine particle fraction. HGMS tests have been conducted on contaminated U.S. Department of Energy soils from Fernald, China Lake, Johnston Atoll, Rocky Flats Plant (RFP), Idaho National Engineering Laboratory, Los Alamos, and the Nevada Test Site (NTS). These soils or soil washing residues contain plutonium or uranium contamination.

Test results to date on Fernald uranium-contaminated soils indicate that HGMS can effectively reduce the uranium concentration in nearly 75% of the soil mass to below 70 ppm. On NTS plutonium-contaminated soils, HGMS tests concentrated 80% of the plutonium into a much smaller volume. Several HGMS tests were performed on RFP soil residues. In one test, 52% of the plutonium was removed from the initial slurry with about 3% of the bulk mass concentrated in the matrix.

HGMS is also being developed for magnetic filtration of contaminated water streams such as that from the Leadville Mine Drainage Tunnel Treatment Facility. In these cases, magnetic separation of elements that are not intrinsically magnetic is accomplished by the proper seeding of iron-based compounds to the waste stream, which act to scavenge the nonmagnetic contaminants. One embodiment of this technology employs the use of finely divided ferromagnetic particles to intercalate or adsorb contaminants onto the surface thereby allowing adsorbate removal by magnetic separation of otherwise non-magnetic contaminants. Early studies have shown that Cd and Pb ferrites can be generated at low temperatures. It follows that these highly magnetic particles can be easily separated by a magnetic separation process.

5. Summary

In summary, we have shown that magnetic separation can be used to concentrate rare earth oxides or actinides from extraneous materials yielding more efficient recovery and treatment operations. We have demonstrated that the extraction and concentration of paramagnetic actinides (PuO_2, UO_2, etc.) from soils is feasible, where our actinide separation results have shown that HGMS is effective for extracting >90% of small radioactive particles from soil slurries. In addition, recovery of rare earth oxides such as Pr_2O_3, Gd_2O_3, and Ce_2O_3 has successfully been demonstrated by HGMS and MRS. Analytical models were developed that describe both magnetic separation process. The model provides guidance in selecting the appropriate bench scale experiments to perform and assists in analyzing the resulting data. A validated analytical model also supports prototype design and process scaleup. We anticipate that with proper pretreatment, mainly consisting of size reduction, a permanent magnet roll separator will be the method of choice for recovery of rare-earth oxides. While magnetic separation cannot extract diamagnetic products (such as La_2O_3) from the soils, we expect that over 90% of paramagnetic rare earth oxide components can be extracted efficiently and economically.

6. References

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PURIFICATION AND UTILIZATION OF WASTE WATERS FROM SILMET PLANT CONTAINING AMMONIUM SALTS

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The waste waters coming from the rare earth metals production plant of Silmet Ltd. contain a large amount of ammonium salts. These waste waters are seriously polluting the Finnish Gulf. Ammonia is also being emitted into the atmosphere due to contact of the waste waters with alkaline ash from oil shale combustion. The treatment of an ammonium-containing waste water must simultaneously solve two problems: the decontamination of the water and the recycling of ammonia or ammonium salts.

The main process streams containing ammonium from Silmet are the so-called "chloride solution" and "nitrate solution." Both of them result from the precipitation of rare earth carbonates from chloride or nitrate solutions with ammonium carbonate in different stages of the process. We have examined a chloride solution containing 54 kg/m^3 Cl^-, 15 kg/m^3 NH_4^+, 8 kg/m^3 Ca^{2+}, 0.5 kg/m^3 lanthanides, and trace amounts of other components; we have also examined a nitrate solution containing 57 kg/m^3 NO_3^-, 17 kg/m^3 NH_4^+, and less than 5 kg/m^3 other components, including lanthanides.

For the chloride solution, the most probable method of ammonia recycling is alkaline decomposition of ammonium chloride. In this paper, this method is described with local materials (burnt lime and oil shale ash) used as alkaline reagents.

The reaction rate and the yield of ammonia depend on both the temperature and the excess of alkaline reagent, i.e., in our case the amount of active calcium oxide. Ammonia can be separated from the reaction mixture by distillation or bubbling with air or with steam. An aqueous ammonia solution is produced as distillate.

Figure 1 shows the dependence of the removal of ammonia on the nature of the initial solution distilled. The experiments were performed with an excess of alkaline reagent. Curve 1 shows the removal of ammonia by the use of pure calcium oxide (97.5%) at 1.18 times the stoichiometric amount. As can be seen, 64% of the bound ammonia can be removed by evaporating 35% of the solution. The distillate contains 2.4% ammonia. Curve 2 shows ammonia removal by oil shale ash containing 15.7% active CaO at 1.16 times the stoichiometric amount. In this case, the ammonia removal is slightly more complete, and 66.5% of the bound ammonia can be removed by distilling 32% of the solution. the distillate contains 3.1% ammonia. Nevertheless, in both cases the excess of CaO remained too small for complete removal of ammonia.

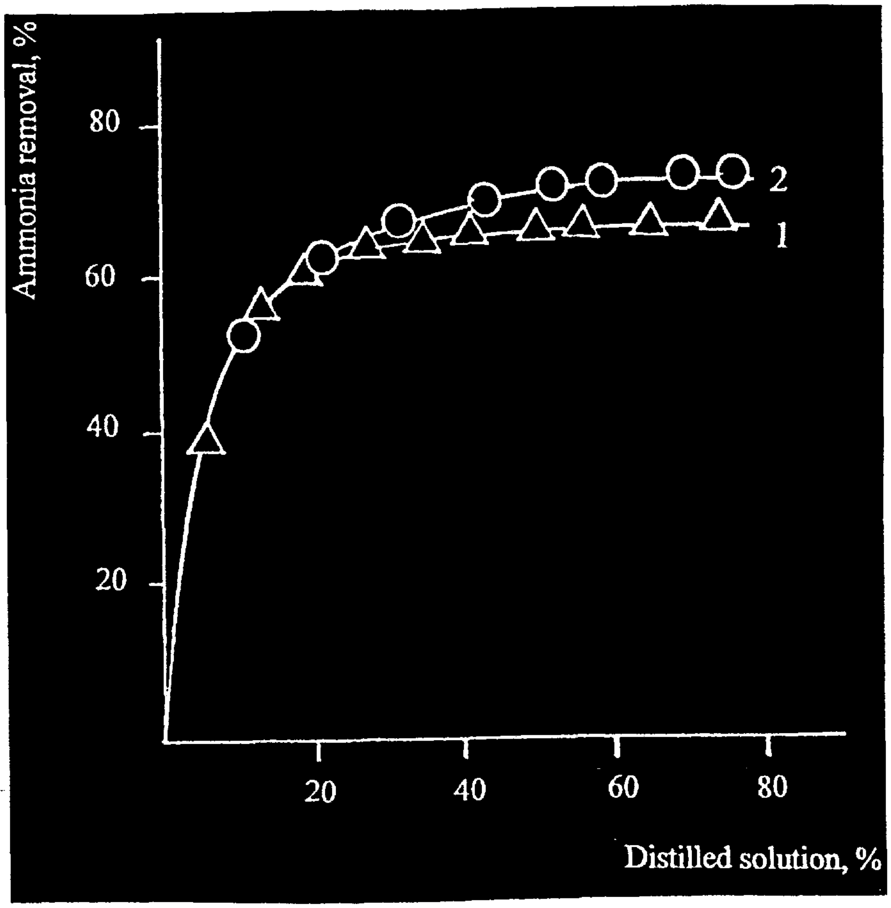

FIGURE 1. Dependence of ammonia removal on the amount of solution distilled. Curve 1: pure (97.5%) CaO. Curve 2: oil-shale ash (15.7% active CaO).

In Figure 2, the results of experiments carried out with a greater excess of alkaline reagent are shown. Curve 1 shows ammonia removal by pure calcium oxide at 1.66 the stoichiometric amount. Curve 2 shows ammonia removal with burnt lime (76.7% active CaO) as the alkaline reagent. The amount of active CaO was 1.58 of stoichiometric. In the case of pure calcium oxide, 81% of all bound ammonia can be removed by evaporating 35% of the initial solution. In the case of burnt lime, ammonia removal remains somewhat lower. An aqueous ammonia solution containing 3.4% ammonia was produced as distillate. A more concentrated ammonia solution can be produced by subsequent rectification.

The slurry from the distillation residue can be filtered to produce a $CaCl_2$-NaCl solution. The concentration of this solution depends on how much of the solution has

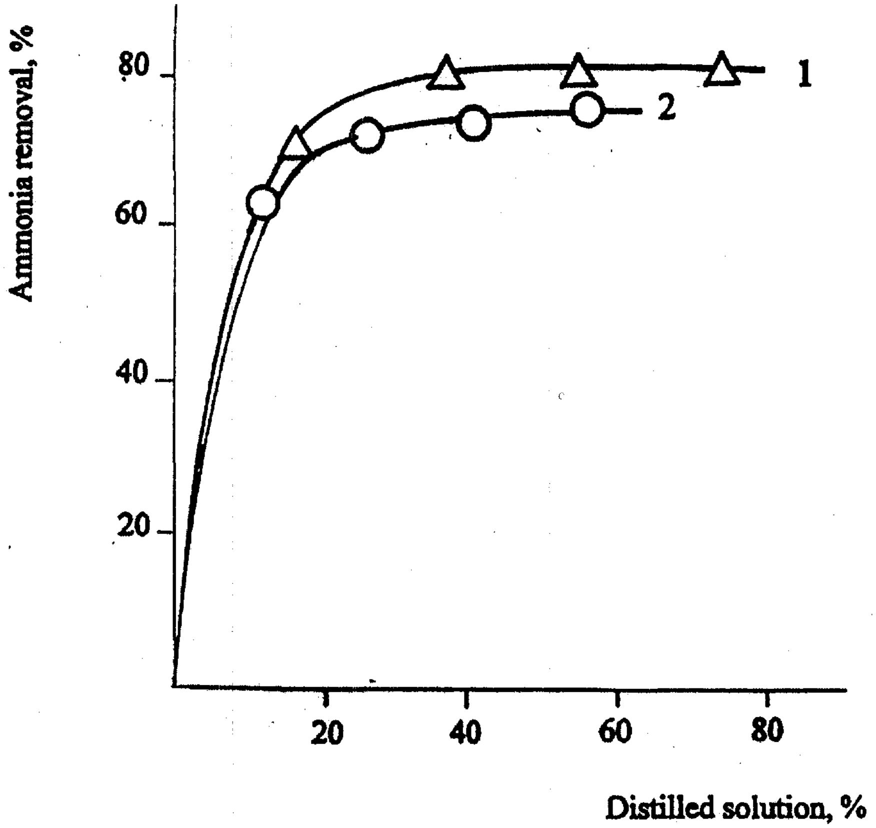

FIGURE 2. Dependence of ammonia removal on amount of solution distilled.
Curve 1: Pure CaO. Curve 2: burnt lime (76.7% active CaO).

been distilled. By evaporating 73% of the solution, filtration of the resulting slurry produces a filtrate containing 20% $CaCl_2$ and 7% NaCl. This solution can be used for preventing icing and dusting on local roads.

The production of a concentrated $CaCl_2$-NaCl solution depends on the costs of evaporation and filtration. A detailed economic analysis taking into account both the production costs and compensation for environmental damage would be necessary. Expenses for the storage and transport of aqueous solution must also be included. Because of economic considerations, the distance from the facility for using the $CaCl_2$-NaCl solution is limited (probably to a radius of about 90 km). The voluminous precipitate remaining after using oil-shale ash as the alkaline reagent is difficult to filter. Therefore, for the production of a $CaCl_2$-NaCl solution, burnt lime is preferred.

The most promising possibility for using the nitrate solution os the production of ammonium nitrate. Ammonium nitrate is widely used in the manufacture of fertilizers and explosives. Crystalline ammonium nitrate can be produced from the nitrate solution by evaporation followed by crystallization and drying. The results of our experiments show the possibility of producing, in a single-step process, a product containing 96-97% ammonium nitrate. Such product purity is sufficient for use as a fertilizer but insufficient for the production of explosives. Higher purity ammonium nitrate can be produced by repeating the crystallization.

As with the chloride solution, the nitrate solution can also be decomposed by an alkaline reagent (lime) to get NH_4OH and $Ca(NO_3)_2$ solutions. The resulting $Ca(NO_3)_2$ solution could be added to the $CaCl_2$-NaCl solution as an agent for inhibiting corrosion.

As mentioned above, a weak ammonia solution can be concentrated by rectification. Nevertheless, the recycling of ammonia from weak aqueous solutions (distillates, absorbates, etc.) is quite problematic. We have demonstrated the ability to remove ammonia from a weak aquesou solution by a gas (air) membrane immobilized within the pores of a hydrophobic microporous filter separating two aqueous solutions (i.e., weak ammonia as the feed solution and sulfuric acid as the stripping solution). Thus, both the desorption of ammonia from water and the absorption by acid solution can be carried out in the same apparatus simultaneously [1, 2]. For the hydrophobic membrane material, microporous polytetrafluoroethylene, polypropylene, polyvinyl chloride, and the perchlorovinyl filter of Silmet Ltd. can be used [2-4].

The mass transfer rate of ammonia in an isothermal unsteady-state batch reactor (in the case of sufficient mixing of both the feed and stripper solutions) can be expressed as follows:

$$\ln(X_{A0}/X_A) = (k_A AH/P_i G_F)t \qquad (1)$$

Where G_F is the molar quantity of the feed; X_{A0} and X_A are the initial and current mole fraction of ammonia in the feed, A is the membrane area, H is the Henry's constant of ammonia, P_i is the partial pressure of the stagnant gas, k_A is the overall mass transfer coefficient of ammonia, and t is time. Equation 1 gives a linear plot of $\ln(X_{A0}/X_A)$ [coinciding with $\ln(C_{A0}/C_A)$ in a weak solution] versus time.

Figure 3 shows both the current ammonia concentration in the feed solution and the ammonia concentration ratio (C_{A0}/C_A) plotted against time. As can be seen, the experimental data points are in good agreement with the linearity of Eq. 1 and also demonstrate a high removal of ammonia. In our experiments, ammonia removal of more than 99% was achieved and the possibility of simultaneously producing a solution of more than 10% ammonium sulfate was shown.

The mass transfer of ammonia can be remarkably accelerated by making use of the temperature difference between the feed and stripping solutions in the reactor. At the same time, ammonia transfer is accompanied by a vaporization of water [4, 5]. The thermally forced membrane desorption of ammonia can be described by

$$X_{A0}/X_A = (G_{F0}/G_F)\exp(k_A H/k_W(P_{WF} - \gamma_W P_{WS}) - 1), \qquad (2)$$

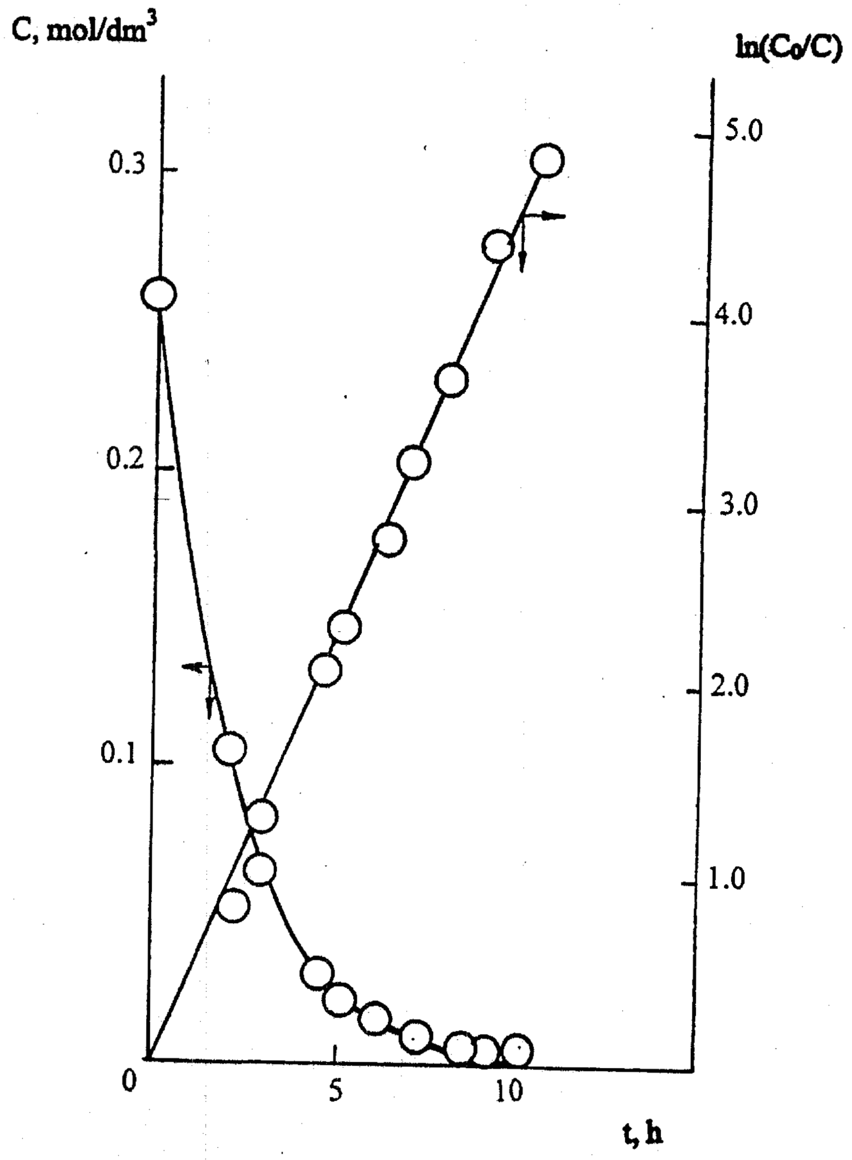

FIGURE 3. Dependence of total ammonia concentration, C, and $\ln(C_0/C)$ on time.

where G_{F0} and G_F are the initial and current molar quantities in the feed, P_{WF} and P_{WS} are vapour pressures of water in the feed and stripping solutions, γW is the activity coefficient of water in the stripping solution, and k_W is the mass transfer coefficient of water.

Equation 2 in logarithmic form gives a family of linear plots, the slope depending on the difference between the temperatures of the feed and stripping solutions as well as on the initial ammonia concentration and molar quantities in the feed. In Figure 4, the experimental data and linear plots of $\ln(X_{A0}/X_A)$ versus $\ln(G_{F0}/G_F)$ at some temperature differences are shown. The experimental data points are in good agreement with Equation 2.

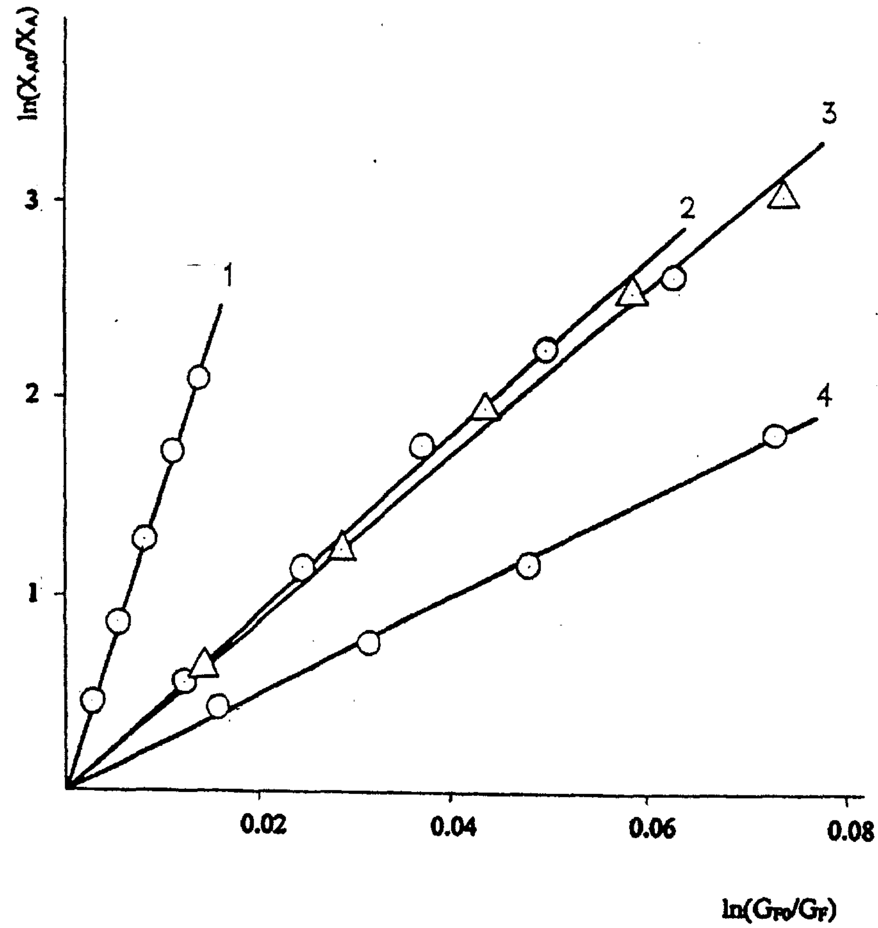

FIGURE 4. Dependence of ammonia concentration ratio on feed mole ratio at elevated temperature differences.
Curve 1: $G_{P0} = 8.31$ mol, $\Delta T = 4.49$ K. Curve 2: $G_{P0} = 8.67$ mol, $\Delta T = 4.00$ K
Curve 3: $G_{P0} = 11.62$ mol, $\Delta T = 11.72$ K. Curve 4: $G_{P0} = 9.34$ mol, $\Delta T = 0.22$ K

Although good results were obtained with gas membranes, some problems connected with their durability will need additional investigation before they can be applied at full scale.

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NEW APPROACHES FOR THE TREATMENT OF NATURAL OCCURRING RADIOACTIVE MATERIALS

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1. Introduction

Over the last several years there has been growing awareness that naturally occurring radionuclides can be an important source of external and internal exposure for workers and the population. Several industrial processes enhance the concentrations of these nuclides in waste streams, which may lead to significant radiation protection problems. An example is the phosphate industry where, depending on the specific process, either gypsum with a relative high content of radium or a calcinate with a high content of lead is set free.

Some of these industries already produce large quantities of waste materials, which exceed their national exemption levels. In general, these waste materials have to be stored in repositories for nuclear wastes. These repositories are very expensive and not equipped for the disposal of large quantities. Even if the storage capacity of the repositories were enlarged, they still represent a very expensive disposal treatment for the industries.

The European regulation regarding the limits for radioactive nuclides in materials was changed in May 1996, and has to be implemented before 13 May 2000 [1]. The new EC exemption levels for natural radionuclides are stricter compared with the existing national exemption levels. Implementations of these exemption levels in the national legislation of EU member states will therefore result in an enlargement of the waste stream characterized by its large volumes and slightly enhanced radioactivity.

Large volumes of radionuclides containing waste has been accumulated during the last decades at the Sillamäe site (Estonia). To prevent dispersal of this waste in the environment, measures have to be developed. As for the radioactive waste producing industries, one of the potential options for the Sillamäe site is to reclaim the waste material by immobilization and bring it to a landfill. The requirements for this immobilization are to control the mobility of the radionuclides and to produce a durable composite with certain strength properties. In this paper a general approach for separation and immobilization of radionuclide containing materials is described as well as the first results of a specific binder waste material combination.

2. Options for solutions

One possible strategy for solution for these types of waste streams might be the immobilization of the radionuclides in a solid matrix in order to store it into alternative nuclear wastes repositories [2]. If it is certain that the radionuclides are stabilized in the matrix and do not reach the environment, even re-use of the materials may be considered as an option. The large volume of the waste streams can also be reduced by separation technologies which produce waste with an increased content of radionuclides and "radionuclide free" waste. Separation techniques can be considered if (i) the concentration of radionuclides is just a few times the value of the Basic Safety Standards (BSS), (ii) the radionuclides are heterogeneously divided in the material and (iii) the activity is linked with specific material properties.

The reduced amount of waste with the high content of radionuclides should then be immobilized for other lower costs alternative disposal facilities. The remaining volume with a radioactive content below exemption levels can be recycled or disposed using standard treatments (Fig. 1).

The most effective choice of treatments of the materials depends on the EC and national legislation, the specific properties of the waste materials, the content of the radionuclides and the half-lifes of the involved nuclides. In addition, economical and logistical aspects should also be considered. The treatment approach of the materials based on the radioactive content and half-lifes, is schematized in table 1.

Technologies for the immobilization of radionuclides in non-nuclear wastes are essential for this strategy. Immobilization technologies can be classified in cold-bonded and thermal treatment processes. In the cold-bonded technology, a mixture of binders and the material, which has to be immobilized, are moulded and hardened. Melting techniques use more complicated installations for the required high temperature and the prevention of various types of emissions [3, 4].

Nuclear wastes are, in general, immobilized in special (costly) containers before storage in repositories [5, 6]. These repositories are very expensive due to the pre-treatment, the particularly long-life time of the wastes and the required health physics facilities. After alternative treatment of wastes with enhanced radionuclide content (Fig. 1) special containers will generally not be necessary and the waste can be moulded in desired shape before storage.

2.1. CRITERIA AND REQUIREMENTS FOR IMMOBILIZATION

Storage of wastes with enhanced levels of natural radionuclides in alternative, less expensive repositories is only reasonable if the radionuclides are chemically or physically stabilized and thus prevented from reaching the environment. In order to test this it is necessary to have (i) a measurement method available for the determination of the leachability of the radionuclides, and (ii) legislation on the permitted leachability. If the

TABLE 1. Treatments and storage of waste materials with enhanced concentrations of radionuclides depending on the activity content and half-lifes.

	Half-life	
content radionuclides	Short	Long
Low (slightly enhanced: 1-3 times BSS level)	Separation and/or Immobilization and/or Temporary storage on plantsites	Separation and immobilization
High	Immobilization	Nuclear waste repositories

leachability of the radionuclides exceeds the permitted levels, immobilization techniques can be used to reduce the leachability. A test method for the determination of the leaschability and the existence of leachability limits are thus essential factors for the development of appropriate immobilization techniques for radionuclides in non-nuclear wastes. Until now neither test methods nor accepted limits have been available. This regulatory framework has yet to be developed and quantified according to the risks for the ecological system and workers.

A regulatory framework and standard test have been developed for non-radioactive inorganic components in soil, stony building material or wastes in the Netherlands and other EU-member states [7]. Because of similarities between the inorganic compounds and radionuclides it is likely that the development of a regulatory framework and standard test for radioactive inorganic compounds can follow a similar route. During this stage it is expected that adaptation is necessary for radioactive nuclides in waste material.

The allowable limits of leachability of radionuclides (in produced immobilisates) could be based on limits of soil contamination with radionuclides. In standard leachability tests several main factors, like pH and liquid/solid ratio, are set on rather extreme values in order to shorten the test period, and thus only partly simulate reality [8]. Therefore, a procedure has to be developed in which the release of different radionuclides in the environment over a long-term period can be simulated. Most natural radionuclides are heavy atoms, or even heavy metals and similarities between the leaching behaviour of heavy metals and radionuclides are thus likely. Some relevant differences between the leaching of radionuclides and heavy metals have, however, to be taken into account. The most important are: (i) the involved concentrations (% by weight) of radionuclides in the leachates can be extremely low, (ii) the ingrowth and decay of radionuclides in the material and leachates result in fluctuations in concentration in material as well as in leachates and (iii) the unknown long-term effect of radiation damage on the leachability. These aspects makes the interpretation of the results of leaching tests more complex compared with "standard" leaching tests.

2.2. REUSE OF IMMOBILIZED MATERIAL

Besides the possibility of a cheaper deposal of large amounts of wastes, immobilisates of this material could be reused. The immobilized material could be applied in the building industry or in the infrastructure works (roads, dams and dykes) or used as back-fill of old mining shafts. For reuse, the immobilisates have to meet additional requirements depending on the application. For example the necessary strength needed in some back-fill applications is about 5 - 10 MPa. When used in road building or dams the demands on the strength can be even higher. Also durability aspects like freeze-thaw resistance can be a very important parameter. Reuse could only be considered if the costs of necessary pretreatment can be balanced with the economical benefits of the application. As an indication the treatment described in this paper costs ca. NLG 80-120 per ton of waste material. This is a potentially interesting option, compared to landfill prices in the Netherlands.

3. Materials and methods

3.1. PRELIMINARY EXPERIMENTS

A research programme has been set up as a first step to develop a measurement technique for the determination of the leachability of radionuclides. As a first step, the availability of radionuclides from fly ash has been determined in order to test the standard procedure for inorganic compounds. In addition, a leachability test was conducted on two types of immobilisates.

3.2. TEST OF AVAILABILITY

Samples of fly ash with enhanced concentrations of natural radionuclides were exposed in conformity with the Dutch standard for the determination of the availability of inorganic compounds to leaching [9]. The availability is defined as the quantity of a certain component, which can leach out of a material under an aerobic environment and relatively extreme conditions (such as disintegration of the material, full oxidation and/or elimination of the acid-neutralization capacity). In this test, very fine-grained material ($< 125\ \mu m$) was mechanically shaken during 4 hours at pH=7 and at pH=4. pH was adjusted with nitric acid and the liquid/solid (L/S) ratio was between 2-10. The extractions were analysed on radioactive nuclides. This procedure was done with an increased involved volume of the material to maintain radionuclide concentrations in the leachates below detection limits. By measuring the nuclide contents of the material before and after the testing period, the results are controlled with a nuclide balance [10].

3.3. TEST OF LEACHABILITY

Natural sand with high levels of natural radioactivity was pulverized, mixed with cement and additives and moulded. The mix proportions were 75 wt-% of the natural sand and 25 wt-% of (two types of) binder (sample 1 and 2). After a hardening period (> 28 days), the immobilisates were exposed in accordance with the Dutch leachability standard [11]. The two immobilised samples were kept in distilled water during 64 days. The water was refreshed 8 times and sampled on several times after the start of the experiment. In this experiment the eluates were analyzed on potassium (^{40}K) and nuclides within the uranium and thorium series.

4. Results

4.1. AVAILABILITY

The availability of uranium in the fly-ash samples varied between <0.008 and 0.066%and for thorium between < 0.0.03 and 0.013%. The radium availability fluctuated between < 0.2 and 0.6% (Table 2). The measured values for ^{238}U availability (L/S=2-10) were clearly lower than the availability (0.1-1.0%, L/S=100) of the element uranium in fly ashes, as determined by van der Sloot *et al.* [8]. The availability of radium in our fly-ash samples were more or less in the same range (0.1-5%) as those quantified for the chemically similar barium [8, 12].

Preliminary estimates of the radionuclide balances of the fly-ash samples indicate some missing amounts (5-12%) of radionuclides. At this moment it is not clear whether this is caused by the preliminary methods of the determination of the nuclide balance or by the unexpected processes which lead to losses of radionuclides. Further research is necessary to adjust and validate the test set-up in this respect.

4.2 LEACHING

The total amounts of radionuclides in the leachates were very low. With gammaspectrometry most nuclides were below the detection limits. The contents of radionuclides which could be measured are shown in Table 3. The interpretation of the results is complicated due to the very low amounts of radionuclides (low reliability) and the mentioned complexity. The determined leaching percentages of the radionuclides and eventually the leached parents are, however, of the same magnitude as has been found for the leachability of non-radioactive uranium of 0.01-0.05% by Van der Sloot *et al.* [8]. In addition, a consistent difference in leaching of the measured radionuclides from sample 1 and 2 has been found (Table 3). The leaching from sample 2 was twice as high as that from sample 1. This indicates an effect of the type of binder on leaching from test samples.

TABLE 2. Availability (%) of the radionuclides ^{238}U, ^{232}Th and ^{226}Ra in fly-ash samples

		Availability	
fly-ash sample	^{238}U	^{232}Th	^{226}Ra
1	0.026 ± 0.008	< 0.006	< 0.6
2	< 0.02	< 0.005	0.6 ± 0.3
3	< 0.008	< 0.005	0.3 ± 0.2
4	0.066 ± 0.016	< 0.003	< 0.5
5	0.039 ± 0.005	0.013 ± 0.003	< 0.2
6	< 0.008	0.005 ± 0.002	0.2 ± 0.1

TABLE 3. The amount of radionuclides (Bq) in the sand samples and their leachates

nuclide	^{214}Pb (U-serie) (Bq)		^{214}Bi (U-serie) (Bq)		^{212}Pb (Th-serie) (Bq)		^{40}K (Bq)	
Sample	1	2	1	2	1	2	1	2
Activity in sample	1725	1440	2157	1800	400	333	?	?
Activity in leachates after								
Step 1 0.25 d	0.07	0.2	0.2	<0.27	0.07	0.1	<2.3	<3.3
Step 2 1,0 d	<0.23	0.1	<0.28	0.19	0.15	0.31	0.5	2.1
Step 3 2.,25 d	0.09	0.15	0.06	0.31	0.23	0.18	1,2	2.2
Step 4 4 d	0.29	0.25	0.62	0.51	0.32	0.86	<2.1	1.7
Step 5 9 d	<0.23	0.22	<0.29	0.29	<0.21	0.17	<2.0	2.5
Step 6 16 d	0.11	0.19	0.13	0.37	0.03	0.25	0.19	0.7
Step 7 36 d	<0.3	0.07	<0.3	0.12	<0.22	0.07	<2.6	4.2
Step 8 64 d	0.22	<0.29	0.24	<0.32	0.54	0.09	<1.9	0.8
Total amount of leached nuclides	0.78-1.54	1.18-1.47	1.25-2.12	1.79-2.38	1.34-1.77	2.03	1.89-12.8	14.2-17.5
Leaching %	0.04-0.09	0.08-0.10	0.06-0.10	0.1-0.13	0.34-0.44	0.6	?	?

5. Conclusions

To restrict the quantities of radioactive wastes from non-nuclear industries, which have to be stored in repositories for nuclear wastes under present legislation, new legislation on the leaching of radionuclides must be developed. Within this regulation the leachability framework for inorganic components can be used as a base for the development of tests for the leachability of radionuclides. Specific factors, however, must be either adjusted or added to this framework.

Preliminary results showed that most concentrations of the radionuclides in the leachates were below or just slightly above the detection limits of the method used. However, these low concentrations do not necessarily indicate that there are no risks involved with this rate of leaching. In cases where the leachate rate exceeds the risk limits, the detection limit has to be lowered. This could be lowered by either using other detection methods and/or adaptations of the procedure.

From the nuclides concentrations which could be determined it can be concluded that:

- the method used, though altered, gives comparable results as the standard method for inorganic compounds;
- some differences have been found between two cold-bonded immobilization types, which suggest that immobilization can be used to decrease the leachability of radionuclides.

Based on the composition of the Sillamäe waste, it is expected that similar strength and durability properties can be reached using this cold-bounded immobilization method. Immobilization could also be an economical solution for the Sillamäe problem, depending on local price levels for landfill and logistics.

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ASH CEMENTS STABILIZED BY SUPERCRITICAL CO_2 CARBONATION FOR TAILINGS POND OVERLAYER

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1. Oil Shale Ash

Estonia possesses large oil shale deposits. The kukersite shale, located in the northeastern part of the country, is arguably the most important mineral resource of Estonia. There are two principal deposits; the *Estonian* and the *Tapa*. The former has a productive seam thickness that varies from 2.7 - 3.0 m in the northern part of the deposit to 1.4 - 2.0 m in the southern and western parts. The Tapa deposit is situated southwest of the Estonian deposit with a seam depth of 60-170 m below the surface. This seam has a maximum thickness 2.0 - 2.3 m in the central part of the deposit. This deposit is not currently in use. These two deposits cover a combined area of about 5000 km^2.

Approximately 80% of the mined oil shale is used to produce electricity by direct combustion in power plants. Unfortunately, since oil shale is composed largely of inorganic mineral matter, this activity generates a large amount of residue ash, amounting to around 5 million tons annually. At present, the most effective way of using this shale ash is as a feedstock in the production of cement. The finest fraction corresponds in composition and binder properties (Table 1) to the burnt oil shale specified in DIN 1164, Part 100. By inter-grinding 20 to 30% of this ash with cement clinker it is possible to produce a portland-oil shale cement (CEM II/B-T 52,5) which hardens quickly and has pozzolanic properties [1]. More than 5 million tons of this cement have already been produced. Reinforced and pre-stressed concrete structures have been built using this cement, including bridges, chimneys and the Tallinn television tower. Despite this re-use, however, stockpiles of spent shale are rapidly accumulating.

TABLE 1. Properties of different size fractions of oil shale ash[2]

Property	Fine fraction	Finest fraction
Specific surface area (m^3/kg)	50-120	320-500
Particle size (μm)	30-90	10-30
CaO	46-58 %	28-35 %
SiO_2	20-28 %	30-35 %
Al_2O_3	6-8 %	10-12 %
K_2O	1-2 %	4-6 %

We have shown that accelerated carbonation of portland cements, using supercritical CO_2, can greatly increase the fraction of ash that can be incorporated into the cement mix. Further, the mechanical and chemical properties, as well as the leachability, of supercritical CO_2-treated portland cement are superior to untreated portland cement. In this paper, we describe the general features of supercritical fluids, as well as the application of these fluids to the production of unmodified portland and portland cement modified by incorporation of industrial waste solids, such as oil shale ash. Suitably treated, pre-fabricated cement, incorporating spent oil shale ash, as well as some of the tailings pond waste, could be used as an overlayer for the Sillamäe tailings pond.

2. Supercritical Fluids

The temperature at which the vapor pressure above a pure liquid reaches one atmosphere is known as the normal boiling point. For water, the normal boiling point at one atmosphere is 100°C. In an open container, Figure 1, the temperature of liquid water cannot be raised above 100°C since this would cause the vapor pressure of the water to rise above one atmosphere, which would exceed the ambient pressure conditions. If we place a quantity of water in a sealed container, however, then we may heat the liquid water to higher temperatures, since the vapor pressure of the water can increase beyond one atmosphere. As we uniformly heat the sealed container, the density of the liquid water decreases through thermal expansion. Simultaneously, the density of the water vapor increases. We can continue this heating process until, eventually, the density of the liquid becomes so reduced, and the density of the vapor phase is so increased, that the two densities become equal. The temperature at which the liquid and vapor densitiesbecome equal is called the critical temperature. Since the temperature inside the sealed container is everywhere equal, and the density is everywhere equal, thermodynamics dictates that the pressure inside the container be everywhere equal. This pressure is called the critical pressure. A fluid that has been brought to conditions above its critical temperature and pressure is known as a supercritical fluid. This physical description of the critical conditions suggests that all simple liquids (and gases) can be made into supercritical fluids by generating the appropriate conditions of temperature and pressure. This is indeed correct, and Table 2 gives the critical temperature and pressure of some common fluids.

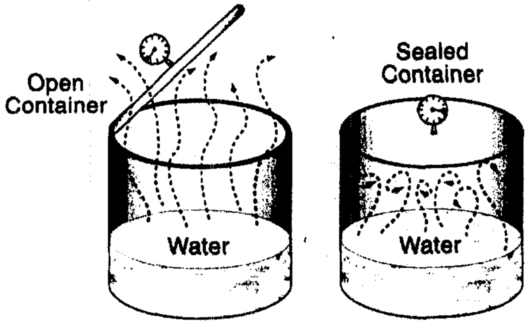

FIGURE 1. Formation of a supercritical fluid by heating a liquid in a sealed container.

TABLE 2. Critical temperature and pressure for some common fluids.

Fluid	Critical temperature (°C)	Critical Pressure (psi)
Neon, Ne	-229	400
Nitrogen, N_2	-147	492
Argon, Ar	-122	706
Xenon, Xe	17	858
Carbon dioxide, CO_2	31	1072
Sulfur hexafluoride, SF_6	46	545
Propane, C_3H_8	97	617
Ammonia, NH_3	133	1654
Water, H_2O	374	3209

Supercritical fluids are used as solvents in many commercial applications, including the extraction of caffeine from coffee and essential oils and spices from plants for use in perfumes and foods. The attractiveness of supercritical fluids as solvents stems from their unique combination of liquid-like and gas-like properties. A comparison is given in Table 3 of the diffusivity, viscosity and density of a typical organic fluid in the liquid, gas, and supercritical fluid state. To a first approximation, the solvent power of a fluid is related to its density. The high, liquid-like densities achievable in supercritical fluids therefore allow for substantial solubilities. Figure 2 shows the pressure-temperature-density surface for pure CO_2. The critical point for pure CO_2, 31°C or 88°F and 1072 psi or about 73 atmospheres, is shown in Figure 2 by the solid black circle. It can be seen that relatively small changes in temperature or pressure, near the critical point, result in large changes in density. It is this tunability of density, and therefore tunability of solvent power, which is one of the most attractive attributes of supercritical fluids. Also, the gas-like properties of low viscosity and high diffusivity of supercritical fluids provides for effective mass transport into granular and micro-porous matrices, such cements. Finally, the absence of surface tension provides for excellent wettability.

TABLE 3. Comparison of physico-chemical properties of a typical organic fluid in the liquid, gas, and supercritical fluid state.

	Diffusivity (cm^2/s)	**Viscosity (cP) or ($mN{\cdot}s/m^2$)**	**Density (kg/m^3)**
Liquid	10^{-5}	10^{0}	1000
Supercritical Fluid	$10^{-3} - 10^{-4}$	$10^{-1} - 10^{-2}$	500
Gas	10^{-1}	10^{-2}	1

Supercritical CO_2 occurs naturally in the headspace above oil reservoirs, and high-pressure CO_2 has been used for many years by oil producers in secondary and tertiary recovery processes. Supercritical CO_2 is an excellent choice for large-scale treatment processes for several reasons. CO_2 is non-toxic, non-flammable, and inexpensive. Further, the critical conditions of temperature and pressure are easily achievable in existing process equipment. We suggest that

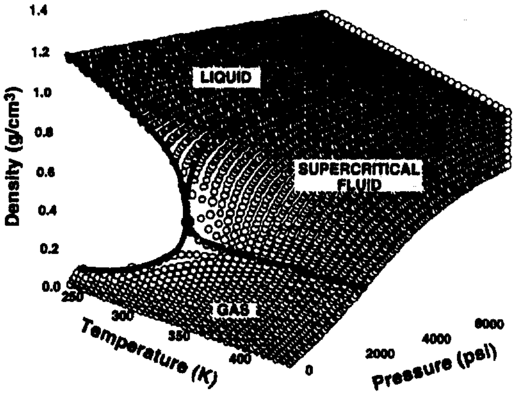

FIGURE 2. Pressure-temperature-density surface of pure CO_2. The critical point is shown as the solid, filled circle.

this unique combination of chemical, physical and economic properties of supercritical fluid CO_2 could be used to produce an ash modified portland cement, useful as a overlayer for the tailings pond.

3. Application to Unmodified Portland Cement

The natural curing reactions that occur in a standard portland cement, Figure 3, involve the formation of portlandite, $Ca(OH)_2$, and calcium silicate hydrates, CSH. Over time, a cured cement absorbs carbon dioxide (CO_2) from the air, converting the portlandite and some of the CSH to calcium carbonate, $CaCO_3$. It turns out, however, that this carbonation reaction results in the blockage and/or closure of pores, impeding the ingress of reactants (CO_2) and the egress of reaction products (H_2O), drastically slowing the reaction rate with time. By exposing a portland cement to supercritical CO_2, the carbonation reaction can be greatly accelerated. This acceleration is due to (1) the ability of the supercritical fluid to penetrate into the pores of the cement, providing continuous availability of fresh reactant in hyperstoichiometric concentrations; and (2) the solubility of the reaction product in the supercritical fluid, facilitating its removal. By accelerating the natural carbonation reaction, a chemically stable product is formed having reduced porosity, permeability and pH, while at the same time significantly enhancing the mechanical strength. The large amounts of $CaCO_3$ that are formed during the carbonation, because of its excellent cementing properties, allow us to replace a large fraction of the portland cement with other materials, such as oil shale ash. Portland cement, modified with ash, displays distinct advantages as an immobilization matrix for heavy metals and radionuclides, and can serve, as in this case, as a low-cost building material.

Manufacture of Cast Cement

$$2(3CaO\cdot SiO_2) + 6H_2O \longrightarrow 3CaO\cdot 2SiO_2\cdot 3H_2O + 3Ca(OH)_2$$

$$2(2CaO\cdot SiO_2) + 4H_2O \longrightarrow 3CaO\cdot 2SiO_2\cdot 3H_2O + Ca(OH)_2$$

$$3CaO\cdot Al_2O_3 + 6H_2O \longrightarrow 3CaO\cdot Al_2O_3\cdot 6H_2O$$

$$4CaO\cdot Al_2O_3\cdot Fe_2O_3 + 10H_2O + 2Ca(OH)_2 \longrightarrow 3CaO\cdot Al_2O_3\cdot 6H_2O + 3CaO\cdot Fe_2O_3\cdot 6H_2O$$

Carbonation with Supercritical CO_2

$$Ca(OH)_2 + CO_2 \xrightarrow{\text{Supercritical } CO_2} CaCO_3 + H_2O$$

$$3CaO\cdot 2SiO_2\cdot 3H_2O + 3CO_2 \xrightarrow{\text{Supercritical } CO_2} 3CaCO_3 + 2SiO_2 + 3H_2O$$

$$3CaO\cdot Al_2O_3\cdot 6H_2O + 3CO_2 \xrightarrow{\text{Supercritical } CO_2} 3CaCO_3 + Al_2O_3 + 6H_2O$$

$$3CaO\cdot Fe_2O_3\cdot 6H_2O + 3CO_2 \xrightarrow{\text{Supercritical } CO_2} 3CaCO_3 + Fe_2O_3 + 6H_2O$$

FIGURE 3. (Top) Idealized chemical reactions occurring during the manufacture of a cast portland cement. (Bottom) Idealized chemical reactions occurring during the accelerated carbonation using supercritical CO_2.

Figure 4 shows the x-ray diffraction (XRD) pattern for a 6" × 12" cement cylinder that was treated with supercritical CO_2 for a length of time sufficient only to produce a reaction in the outer portion of the cylinder. It can be seen that the pattern for the outer portion of the cylinder indicates nearly complete conversion of $Ca(OH)_2$ to $CaCO_3$, while the pattern for the inner portion shows minimal conversion. We observe that the supercritical CO_2 treatment results in carbonation, whose depth depends on the porosity, permeability, and free-water content of the cement prior to treatment. By suitable adjustment of these parameters, we have demonstrated complete carbonation in cement samples during a treatment time of several hours. It is important to note that supercritical CO_2, while solubilizing organic compounds and water, does not leach the metals, as these are incorporated into the crystal structure of the inorganic phases [3].

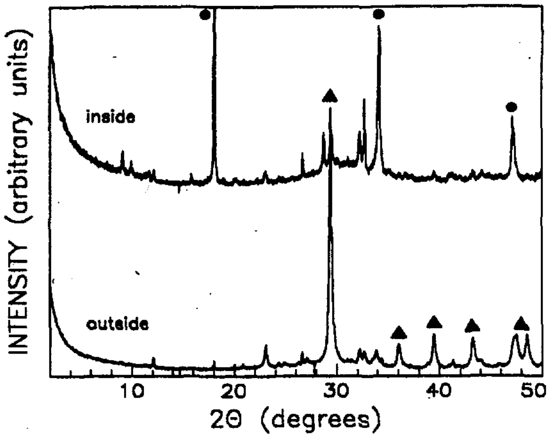

FIGURE 4. X-ray diffraction spectra of the inside and outside portion of a 6" x 12" portland cement cylinder partially reacted with supercritical CO_2. The filled circles identify the peaks due to $Ca(OH)_2$, while the filled triangles show the diffraction peaks due to $CaCO_3$.

We have conducted supercritical fluid treatment experiments on cylindrical cement samples up to 6-in. diameter by 12-in. length, where nearly complete carbonation (water removal) is achieved in a treatment time of several hours. Figure 5 shows permeability measurements for treated and untreated portland cements as a function of curing time. The samples had high initial densities and low initial porosities, so that the carbonation was limited to the outer “skin” portion. Nevertheless, the data clearly show that significant enhancements can be achieved. We find generally that the supercritical CO_2 treatment also results in an increased density and increased compressive strength. Further, the strength enhancement improves with the total curing time, indicating that the maximum strength enhancements will be achieved for cements that have been allowed to fully cure for a period of several weeks. However, steam curing can accelerate the curing process itself. This can easily be accomplished in the same pressure vessel used for the supercritical fluid treatment. An example of the chemical and structural changes produced by supercritical treatment of unmodified portland cement are shown in Figures 6 and 7.

4. Application to Fly Ash Modified Portland Cement

We have been investigating two broad areas of application for portland cement incorporating industrial waste, such as ash, and its treatment using supercritical CO_2. The first is the enhancement of cement, which is itself an industrial waste. By this we mean cements used to immobilize environmentally regulated metals and radioactive elements. In this regard, the tailings material at Sillamäe could be used directly in the modified cement. The advantage of this approach is that the cement could be made on site, using

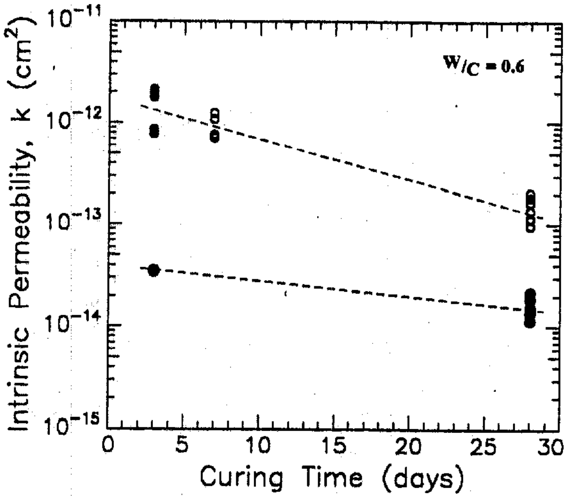

FIGURE 5. Permeability of untreated (open circles) and supercritical CO_2-treated (filled circles) portland cements, as a function of curing time. W/C refers to water/cement ratio of the cement mix.

FIGURE 6. Micrograph of unmodified portland cement before supercritical CO_2 carbonation.

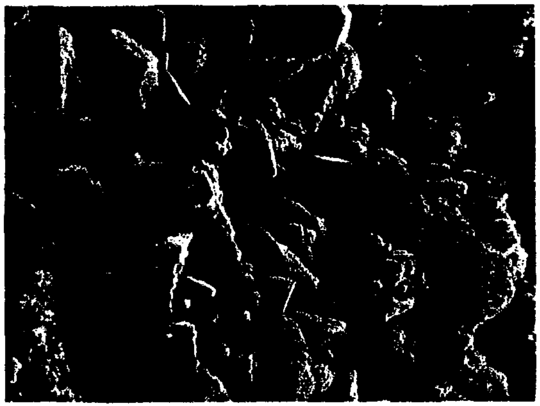

FIGURE 7. Micrograph of unmodified portland cement after supercritical CO_2 carbonation.

oil shale ash brought to the site, mixed with tailings having the highest concentration of metals. There is already an extensive experience database on the performance of fly ash-modified portland cement for radioactive waste immobilization. The United Kingdom [4]and the United States [5] have used these materials, in the form of cement grouts, for the solidification/stabilization of low- and intermediate-level radioactive wastes. In the bulleted text that follows, we review the known benefits of fly ash-modified portland cement over unmodified portland cement, along with the anticipated improvements expected by the supercritical CO_2 treatment of modified portland cement.

- Fly ash increases the density, decreases the permeability, and increases the leaching resistance of OPC. It is a truism that "The leach resistance of solidified cement-waste systems can be improved by any process which accelerates curing, limits porosity, or chemically bonds fission product or actinide elements."[7]. $SCCO_2$ treatment of a modified portland cement is expected to further increase the density over the untreated material, so that reduced porosity and improved leachability should result. In addition, high silica ash, with its well-known sorbent properties towards actinides and certain other radionuclides, can further enhance immobilization characteristics (Table 4).

- The presence of heavy metals is often observed to significantly interfere with proper setting of portland cement. The incorporation of fly ash into portland cement helps to partially compensate for this set-retardation, and a subsequent supercritical CO_2 treatment of such a modified cement should perform even better.

- Fly ash incorporation improves the fluidity of a portland cement mix, which improves workability. For solidification/stabilization applications, this increased fluidity may allow a reduced water/cement ratio to be used in the casting operation.

- The incorporation of fly ash lowers the initial heat evolution during setting, reducing the incidence of cracking and spalling. It is desirable to maintain the modified cement in monolithic form for optimum leach resistance.

TABLE 4. Modified portland cement compositions for the immobilization of heavy metals [6].

Metal	Good	Better	Best
lead	Type I cement	Type I cement with silica fume	Type V with silica fume and a little extra gypsum
cadmium	Cement with silica fume	Higher cement factor, no silica fume	Still more cement for long-term durability
chromium	Portland cement	Portland cement with silica fume	Same but with oxidation inhibitor to avoid hexavalent chromium
arsenic	Portland cement with added $FeSO_4$	Portland cement with pre-oxidation with H_2O_2 and added $FeSO_4$	Same, but with higher cement factor and higher Fe/As ratio
mercury	Portland cement	Higher cement factor	Even more cement to ensure strongly reduced porosity

- Fly ash, as it is typically generated in a high free-carbon environment, contains iron in a reduced state, which helps to lower the redox potential in the cement. Maintaining both heavy metals, such as Cr, and radionuclides in lower oxidation states should result in lower solubilities and reduced leachabilities.

- Since the $SCCO_2$ treatment converts the alkaline phases into neutral pH phases, a straight or fly ash-modified portland cement which has been carbonated will have a much reduced pH. In portland cement, the highly alkaline environment precipitates actinide elements as hydroxides, rendering them immobile. We have recently begun experiments to evaluate the effect of supercritical CO_2 treatment, and the attendant pH reduction, on the leachability of actinide surrogates. We have shown that the treatment improves the leachability, by incorporating the surrogate elements within carbonate phases and in de-calcified regions adjacent to these carbonate phases.

- While portland cement is considered to be an inexpensive immobilization matrix, relative to other candidate materials, its cost is expected to rise in light of current and future projected shortages. Fly ash, as it is a large-volume industrial waste, is both cheap and abundant, so that there is an economic incentive to use fly ash-modified cements. In addition, CO_2 is also produced as a waste by-product of industrial processes (power generation, cement manufacture, etc.), and its permanent sequestration into cement is an added environmental benefit. A fully carbonated portland cement permanently sequesters about 130 liters of CO_2 per kilogram of cement.

The second general area of application that we have been investigating is the utilization of solid wastes, generated as by-products of industrial processes, as additives to conventional cements to be used as low-cost building materials. Examples of such waste materials are fly ash from coal-fired power plants and ash from oil-shale-fired power plants. Because the $CaCO_3$ formed during the accelerated carbonation reaction is itself chemically stable and an excellent cementing agent, it is possible to replace a large fraction of the portland cement with fly ash, while maintaining similar levels of durability and mechanical strength. We have produced molded structures such as roofing tile and wallboard in which the fraction of fly ash is nearly 80 % by volume. Figures 8 and 9 show the profound structural and chemical modifications produced in cemented fly ash microspheres as a result of supercritical CO_2 treatment.

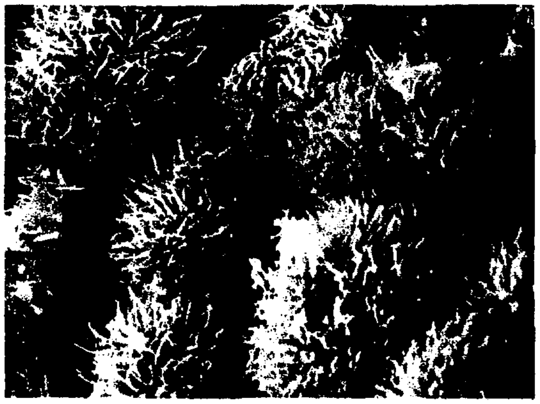

FIGURE 8. Micrograph of cemented fly ash microspheres before supercritical CO_2 treatment.

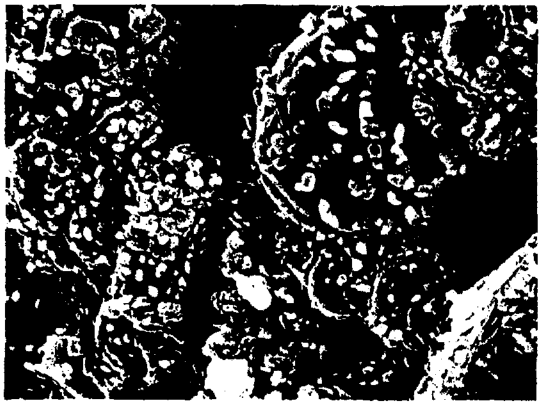

FIGURE 9. Micrograph of cemented fly ash microspheres after supercritical CO_2 treatment.

Using this building materials approach, ash-cement panels could be fabricated at a location near an oil-shale-fired power station and could then be sent to the tailings pond and pieced together to make an overlayer. The advantage of this approach is that the electrical power, heat, ash and CO_2 required for the treatment process could be taken directly from the power station. We are, in effect, using the waste products of the power station to "close the CO_2 loop" on the cement-manufacturing process (Figure 10).

5. Acknowledgements

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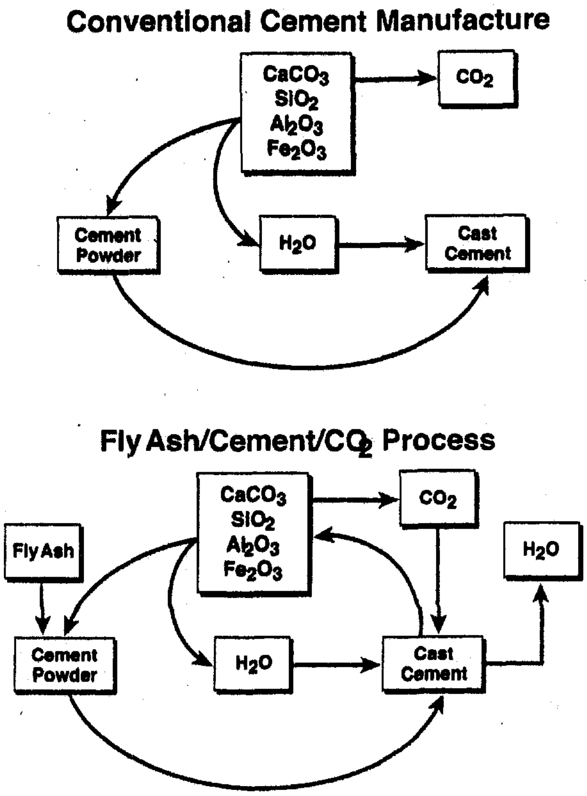

FIGURE 10. (Top) Process flow diagram for the manufacture of unmodified portland cement. (Bottom) Process flow diagram for the manufacture of ash-modified cement using supercritical CO_2.

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A NOVEL CHEMICAL NITRATE DESTRUCTION PROCESS

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1. Introduction

The Sillamäe hydrometallurgical ore processing plant has been in operation since 1948. During the Soviet era, the plant produced uranium as well as rare earths and rare metals. Since Estonia's independence, the rare earth metal processing continued at a smaller scale. A tailings pond was established in 1959 on the beach of the Baltic Sea. The pond collects the waste water and slurry from the plant. Water permeates through the pond and into the nearby Gulf of Finland. Naturally this location makes it a matter of concern for all Baltic countries [1].

Chemical contamination from the pond in the form of nitrates may have significant impact on the ecology of the Baltic Sea. Nitrates, because of their high solubility, are not stopped or filtered by the dike material. They flow freely into the sea. Roughly 10 m^3 of nitrates at a concentration of 40–50 g/l are generated per ton of raw material processed. The production capacity of the plant is a few thousand tons per year and the plant now operates somewhat under that capacity [2].

Nitrate wastes are harmful to humans and the environment if not disposed of properly. Nitrates are especially dangerous for nursing mothers and infants, causing the so-called "blue baby" disease. Additionally, nitrate wastes in a natural body of water can cause excessive biological activity, vegetation growth, and precipitation of organic residue to the bottom. Excessive nitrate concentrations can transform healthy bodies of water into decaying marshes.

A. Siinmaa and V. Petrenko [3] presented four ways to remove nitrates from the plant effluent: (1) producing fertilizers, (2) producing explosives, (3) utilization as nitric acid, (4) denitration using plasma or by burning in a furnace. This paper contains an overview of possible methods of nitrate destruction and presents a novel, non-biological and non-thermal process for converting nitrates to nitrogen gas.

2. Nitrate Elimination Literature Overview

Nitrate destruction methods can be divided in two groups: biotreatment and physicochemical methods.

Biological denitrification involves reducing nitrate to a gaseous nitrogen species. The gaseous product is primarily nitrogen gas (N_2), but it may also be nitrous oxide (N_2O), or nitric oxide (NO). Nitrate can be reduced by a broad range of bacteria. Genera known [4] to contain denitrifying bacteria include *Pseudomonas*, *Micrococus*, *Archromobacter*, *Thiobacillus*, and *Bacillus*. Natural biological denitrification occurs, although not extensively, in aquifers in which a sufficient source of oxidizable organic carbon is present [5]. Water-treatment processes stimulate denitrification by injection of nutrients. Such organic compounds as methanol, methane, glucose, and starch or mixtures (for example, a sugary brewery waste) [5] can be used as carbon sources.

Biological denitrification has been extensively investigated for several reasons: denitrification is a major natural mechanism for loss of fertilizer; denitrification can be used to remove nitrogen from such high-nitrogen waste materials as animal residues; and drinking water resources are facing major nitrate contamination that may cause methemoglobinemia, an infant disease. Biotreatment can remove nitrates down to 2-10 mg/L of total nitrogen [6], potentially below the standard set by US EPA (10 mg/L as nitrate-nitrogen). Some recent reviews are listed [5-7; 10-13].

Biological denitrification is an effective nitrate-removal technology. The disadvantages of biological denitrification are that it is slow, is difficult to control, and produces an organic residue. Bacteria are sensitive to heavy metal ions and to changes in the composition of the influent stream.

Chemical methods for destroying nitrate can be divided into two groups: nonspecific methods and methods designed for nitrate destruction. Seventeen US patents issued since 1976 deal with nitrate destruction. Six patents describe supercritical water oxidation (SCWO) reactors. Eight patents describe thermal processes that achieve nitrate destruction (in six methods nitrates are used as oxidizers; two address thermal nitrate self-destruction). Three patents describe methods for nonthermal nitrate destruction: two of them are electrolytic methods, and one is chemical.

SCWO is a nonspecific nitrate-destroying method. It is based on the use of various oxidizers (for example, O_2, NO_3^-, Fe^{3+}) in supercritical water. The critical temperature and pressure of water are 374 C and 22.9 MPa. Supercritical water is an excellent solvent for gases and most organics. Usually it allows an effective oxidation of organic compounds to H_2O and CO_2; sometimes addition of a heterogeneous catalyst is necessary. Inorganic salts are almost insoluble in supercritical water if the temperature is higher than 425 C. Any phosphorus, chlorine, or sulfur compounds exposed to SCWO are converted into phosphates, chlorides, or sulfates and may be precipitated and separated. SCWO has successfully treated various streams of wastes [14-17]. In some studies, nitrate has been used as oxidizer [16-19]. Wet-air oxidation (WAO) is a similar technology, but it uses slightly lower temperatures (125-320° C) and pressures (0.5-20 MPa) [20]. Both SCWO and WAO are energy-intensive processes and require high pressure. Consequently, special means must be used during treatment of radioactive wastes to modify the hazard of contamination.

Thermal methods of destruction can use the nitrate disproportionation reaction or the nitrate–reducer reaction. The first method may be used to decompose solid nitrates. A general disadvantage of this method is that toxic oxides of nitrogen are produced. The second method is based on using a chemical agent. Cox, Hallen, and Lilga [21] did experiments at 350 C and 2500–2900 psi using six different reducers. The best results were obtained with formate: 100% efficiency of nitrate removal, 99.9% converted to N_2, 0.1% to N_2O. The disadvantages of thermal methods are high temperature and pressure.

Low-temperature, low-pressure methods of nitrate reduction are based on cathodic electroreduction of NO_3^- or chemical reduction.

In the electrochemical methods, electric current is used to reduce nitrate. The ideal process consists of two reactions:

at cathode: $2NO_3^- + 12H^+ + 10e \rightarrow N_2 + 6H_2O$
at anode: $2H_2O - 4e \rightarrow 4H^+ + O_2$.

The real process is more complicated and still not fully understood. Even the first reaction, $NO_3^- \rightarrow NO_2^-$, is a multistep phenomenon with adsorption of different species [19-21]. The mechanism and efficiency of electrochemical nitrate reduction depend on the cathode material. Usually a special catalytic electrode surface is necessary to achieve high efficiency of $NO_3^- \rightarrow N_2$ reduction. The industrial use of the process can be difficult; for example, if metals are present, they may be deposited on the cathode, changing its catalytic properties. The second important factor is nitrite itself; NO_3^- is far less reactive than NO_2^- [22, 23, 25], but it becomes reactive in the presence of NO_2^- [24, 26, 27].

Some attempts [28–30] have been made to use electrolysis to remove nitrate. The composition of N-containing gases produced during electrolysis was found to depend on the electrode material and current density. The best results were achieved [28] with Metcote ES-6 and cadmium cathodes (70–75% N_2; ~25% NH_3; ~2% N_2O), with an overall current efficiency of 55%. The drawback of this method is a low destruction efficiency (64–99% of nitrate destroyed) and high residual NO_3^- concentrations. Even after the best runs, $NO_3^- + NO_2^-$ concentration exceeded 1500 ppm [28].

Two chemical reduction methods have been proposed for nitrate removal. One uses aluminum as a reducer, the second gaseous hydrogen.

In the pH range 9–10.5, nitrate can be reduced to ammonia with aluminum powder [31]. Ammonia can then be removed by air-stripping or by $MgNH_4PO_4$ precipitation. Oak Ridge National Laboratory implemented this idea on a laboratory scale [29]. A bench-scale study [32] with relatively concentrated nitrate (0.1 to 1.0M $NaNO_3$) showed that up to 90% of the nitrate can be converted to ammonia, but residual concentrations of nitrates remain high (~0.03M or ~2000 ppm). An average residual NO_2^- concentration in treated waste was ~1000 ppm. Nitrite is more toxic than nitrate.

Hydrogen can be used as a nitrate scavenger in the presence of a catalyst. Light can be used to initiate the reaction [33]. The best results were achieved with a palladium catalyst [34]; 99.9% of NO_3^- was reduced to nitrogen; only 0.1 % formed ammonia. The drawbacks are that the catalyst is sensitive to poisoning by the presence of heavy metals. The process has been tested only at low concentration of the nitrate.

3. Proposed Process For Nitrate Conversion To Nitrogen Gas

The authors of this article have invented a novel, inexpensive, non-thermal, non-biological, simple process for converting nitrates to nitrogen gas. A patent for the invention is pending. In the process, a metal catalyst is contacted with a nitrate-containing solution or slurry. Nitrates are quantitatively reduced to nitrites while the metal cations are released to this solution. Subsequently an organic reagent is added to the solution and is oxidized by nitrite, which is reduced to nitrogen gas. The dissolved metal cations are electrochemically reduced to metal, recovered, and reused. Figure 1 depicts the process. These steps may be carried out in sequence or simultaneously.

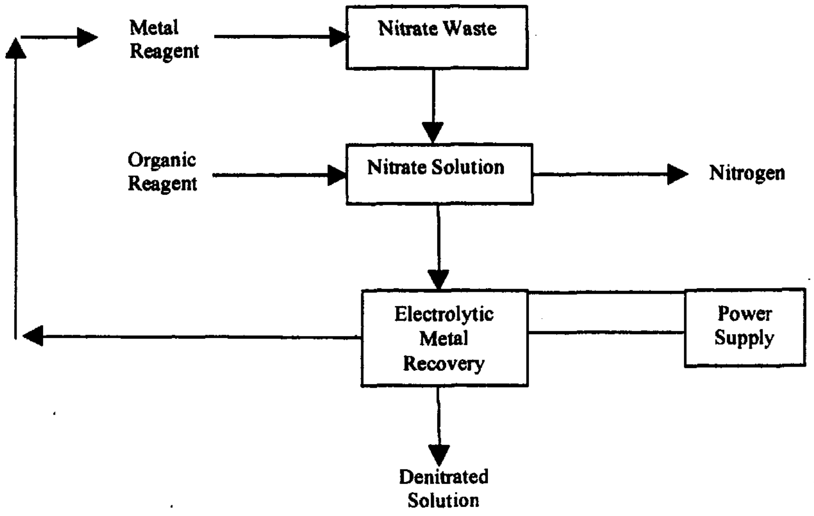

FIGURE 1. Block Diagram of the LANL Nitrate Destruction Process

The process may be arranged in many different ways. It may be carried out in a batch or continuous arrangement. In a batch process, the metallic reagent may be contacted with the nitrate-containing solution in many different ways. For example, a metallic powder may be mixed with the nitrate solution, the nitrate-containing solution may be passed through a bed of dispersed metal, or the solution may be contacted with a solid non-dispersed metal structure, such as metal sheets, spheres or cylinders. Such solid metal structures may be connected to a current source, and metal recovery by electroplating may be carried out in the same vessel as nitrate reduction. The reduction to nitrogen by the organic reagent may be carried out simultaneously with the nitrate reduction or after the nitrate reduction. It may be done in the same vessel as nitrate reduction, or the solution may be pumped to another vessel. When organic reagent has reacted with the nitrite solution to produce nitrogen, the metallic reagent may be recovered electrochemically from the remaining solution. This reaction may be done in a different vessel, or in the same vessel as the nitrogen release reaction. After the three reaction steps are completed, the batch may be released to the environment or to further treatment as necessary.

In a continuous process, the nitrate-containing solution is pumped through a reactor with the metallic reagent. This reactor may be a tank reactor with agitation, or a plug flow reactor with a bed of metal. The metal may be in powdered, dispersed, or solid form. Metal recovery may be performed simultaneously with the nitrate-nitrite reduction step or as a subsequent step. The solution after the reduction of nitrate to nitrite may continuously flow to another reactor where the organic reagent is continuously added. Nitrite will be reduced to nitrogen gas in this second reactor and released from the reactor or captured. Alternatively, the reactor for nitrate-nitrate reduction may also be used for nitrite-nitrogen reduction. In such a scenario, the metallic reagent will be contacted with the influent solution simultaneously with the addition of the organic reagent, and the nitrate-nitrite reduction will be carried out simultaneously with the nitrite-nitrogen reaction. The metallic reagent may be recovered continuously in a separate or the same reactor. The reduced solution containing cations of the metallic reagent may continuously flow to the electrochemical cell. In such a cell the metal will be recovered from the solution by electrochemical reduction on cathodes. The denitrated solution may be released from this cell to the environment, or directed to further processing if required. Alternatively, the electrodes may be in contact with the solution in the same reactor as used for nitrate-nitrite reactor or nitrite-nitrogen reactor and metal recovery performed there.

This process may be used for denitrification of solutions at various nitrate concentrations. For example, it might be used for treatment of radioactive waste water effluents that are high in nitrate concentration, or for remediation of contaminated natural waters with low levels of nitrate. Solid nitrate salts, or heterogeneous solids containing nitrate salts, can be processed by mixing them with water and subjecting to the same treatment.

4. Status Of The New Process

The process has undergone bench scale testing. Initial concentrations of nitrates from 1400 to 200 ppm $N\text{-}NO_3^-$ were successfully reduced to less than 10 ppm. Some of the tests were performed under an argon gas blanket so that the gas produced could be captured for analysis without interference from atmospheric nitrogen. A typical gas analysis is shown in Table 1. It shows that nitrogen was indeed generated with negligible quantities of oxides of nitrogen.

The process is currently being evaluated for implementation at the Los Alamos National Laboratory. Pilot scale testing is in process using a solution simulating the effluent from the Radioactive Wastewater Treatment Plant at Los Alamos. The composition of this solution is shown in Table 2.

The testing is performed with 20-gal batches. A schematic flow diagram for the pilot process is shown in Figure 2. Typical results are shown in Tables 3 and 4. The results show that the process successfully reduces nitrates to nitrogen gas. The gas analysis was made during a test conducted under an argon purge. Some oxygen present in the off-gas may have resulted from air leakage into the reactor tank.

TABLE 1. Gas composition from bench scale testing.

Gas	Mole %
H	0.013
NH_3	<0.05
N_2	78.5
NO	0.2
O_2	<.01
Ar	21.3
CO_2	0.044
N_2O	0.01
NO_2	<0.05

TABLE 2. Simulant Composition Used in Pilot Tests

Analytical Data			Synthetic Waste Composition	
	mg/L	mmoles/L		mmoles/L
SiO_2	100	1.66417	Na_2SiO_3	1.66417
Na^+	800	34.79774	$NaHCO_3$	3.736541
K^+	75	1.918159	$NaNO_3$	14.70378
Ca^{2+}	100	2.49501	Na_2HPO_4	0.484363
Mg^{2+}	40	1.645413	Na_2SO_4	3.258041
HCO_3^-	228	3.736541	NaCl	5.544264
SO_4^{2-}	313	3.258041	$CaCl_2$	2.49501
Cl^-	569	16.05078	$MgCl_2$	1.645413
NO_3^- as N	206	14.70378	KCl	2.225665
F^-	11	0.578947	KF	0.578947
PO_4^{3-}	46	0.484363		
cations		44.99674		
anions		44.70339		
pH	8			
COD	75			
alpha nCi/L	30			
Tritium pCi/L	20000			

TABLE 3. Results of Pilot Testing

	Trial 1		Trial 2	
Component	**Initial**	**After removal**	**Initial**	**After removal**
NO_3	229	0.2	220	2.9
NO_2	0.01	Not detected	0.02	Not detected
NH_3	2	5.65	2	4.9

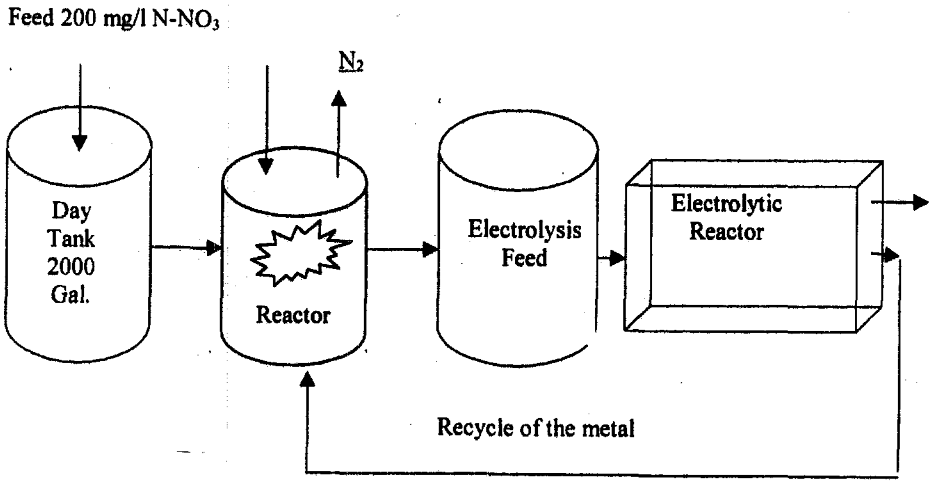

FIGURE 2. Schematic of the pilot plant.

TABLE 4. Gas analysis for pilot tests (Mole %)

Ar	73.9
N_2	23
O_2	2.28
CO_2	0.81
NO	~0.04
H_2	<0.01
NH_3	trace
N_2O	not detected
NO_2	not detected

The process is planned to be implemented at Los Alamos in 1999. The predicted operating costs of the process are $2 per kg of nitrate destroyed. The capital costs for a 2000 gal per day batch operation at 1 g/L of NO_3^- are estimated to be $200,000.

A comparison of the known nitrate destruction processes with our process is given in Table 5.

TABLE 5. Comparison of nitrate destruction methods.

Nitrate destruction method	**Maturity**	**Nitrate concentration in the influent**	**Major nitrate conversion product**	**Byproduct**	**Advantages**	**Disadvantages**
biological denitrification	commercial, industrially applied process	Medium or low	N_2	biomass, NH_3, N_2O	well established, most suitable for non-industrial effluents	sensitive to poisoning and influx changes, slow, secondary waste (biomass)
supercritical water oxidation	bench to pilot scale	Medium to low	N_2	NO_x, NH_3	nonspecific method, a variety of organics and oxidizers may be destroyed at the same time	high pressure, high temperature required, high capital cost
direct electrolysis	bench scale	High	N_2, NH_3	-	room temperature,ambient pressure method, no secondary waste	high residual nitrate concentration (g/L), produces ammonia, difficult to control
reduction with aluminum	bench to pilot scale	high to medium	NH_3	Al hydroxides	simple, low capital cost	produces ammonia, high residual nitrate concentration (g/L), may reduce SO_4^{2-} to H_2S, high reagent costs
reduction with gaseous hydrogen	bench scale	low	N_2	-	suitable for drinking water, no secondary waste	expensive catalyst prone to poisoning, works only with low concentrations
thermal destruction	bench to pilot scale	solid nitrates	NO_x	metal oxides	separates nitrates from solid waste	energy intensive, produces NO_x, may be explosive if organics are present
proposed process	bench scale	high to low	N_2	-	room temperature, ambient pressure, simple, easy to control, broad range of nitrate concentrations, low capital cost, no secondary waste	needs engineering development

High nitrate concentration: 10 - 100 g/L; medium 0.1 - 10 g/L; low - below 0.1 g/L

5. Conclusions

A new non-biological and non-thermal simple process has been recently developed and tested at Los Alamos National Laboratory. This process may be applicable to nitrates in Sillamäe waste water.

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