An Overview of Active and Passive Mine Water Treatment at Urgeiriça Uranium Legacy Mine (Portugal)

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Abstract

Environmental remediation of legacy mining sites in Portugal has been done since 2001 by EDM – Empresa de Desenvolvimento Mineiro, a state-owned company, including radioactive and sulfide polymetallic mines. One of the main focuses of the remediation design projects is the control and treatment of mine water using combined active and passive treatment systems. This paper presents the mine water treatment processes and monitoring that are implemented at Urgeiriça uranium legacy site.

Keywords: Urgeiriça mine, Mine water treatment, Environmental remediation

Introduction

Exploitation of radioactive ores in Portugal initiated with the discovery of the first radium deposit in 1907 and was concluded in the early nineties. Since 2001, EDM – Empresa de Desenvolvimento Mineiro, S.A. (EDM), a Portuguese State-Owned Company, has been granted the responsibility for the environmental remediation of all mining legacy sites in Portugal, under a concession contract established by the Decree-Law 198-A/2001. The implementation of remediation aims for the reduction of risks to people and to the environment and to contribute to the reuse of these areas for different beneficial end-uses, such as restoration of landscapes and habitats, or other public subsequent uses (Carvalho *et al*. 2023).

In the scope of the concession contract are included 199 legacy mining sites, including 62 radium and uranium legacy sites, which have been exploited between 1912 and 1991. Since the beginning of concession, EDM remediated 55 radioactive mining sites and 10 radioactive and polymetallic mining sites are planned to be remediated until 2030.

One of the main focuses of the remediation design projects is the control and treatment of mine water using combined active and passive treatment systems (Diamantino, 2016). This paper presents the mine water treatment processes and monitoring that are implemented at Urgeiriça uranium legacy site.

The treatment system is divided into two lines, active treatment and passive treatment. The active treatment includes pH neutralization with calcium hydroxide and addition of barium chloride followed by sedimentation for solid-liquid separation. The secondary passive treatment system, includes several steps such as aeration, sedimentation, filtration in adsorbent media and phytoremediation in aerobic wetlands.

Both systems are monitored before, in the intermediate steps and at the end of the treatment, and the water quality control programme includes in situ analysis of parameters such as pH, electrical conductivity, temperature, redox potential, total dissolved solids and flow rate and laboratory analysis of chemical and radiological parameters as total uranium, U238, U234 and Ra226 radionuclides, sulfate, chloride, manganese, calcium and sodium. These elements were identified in previous studies as the best indicators of hydrogeochemical contamination related to the legacy uranium mining sites in Centro Region, Portugal (Diamantino *et al*. 2016).

This paper will present an overview on the evolution of the mine water quality and treated volume, including assessment on the effects of the implementation of the remediation of two tailings dams, waste rock piles, uranium mill and other affected areas in the Urgeiriça legacy mining site and the resulting water quality improvement, specific removal efficiency rates and compliance with regulatory limits for water discharge. Also, it will present the quantities of used chemical reagents and operation and maintenance costs of mine water treatment.

Brief description of the Urgeiriça legacy mine site

Urgeiriça mine site is located in the village of Canas de Senhorim, in the municipality of Nelas and district of Viseu, in the central Portugal. Considered at one point to be one of the most important uranium mines in Europe, the Urgeiriça mine was registered in 1912 by a French group for radium exploration. In 1929, ownership of the mine passed into the hands of the Companhia Portuguesa de Radium (CPR) which was formed by English capital. By 1944 exploration restarted, interrupted by the outbreak of World War II, with the production of uranium concentrate. In 1962, the mine passed into the hands of the Portuguese State, through the Junta de Energia Nuclear (JEN), created to promote and develop nuclear energy studies. Around 1977, the National Uranium Company (ENU) took over the management of the mine until its closure in 1991.

Geologically, the Urgeiriça mine is characterized by the presence of porphyroid granites that host the quartz vein being explored, which is 7 kilometers long, 3 to 7 meters thick and oriented N 60º E. It is composed of of quartz, jasper, microbotrioidal pitchblende, uranitite, autunite, torbenite, and sulfides such as pyrite, marcasite, galena and chalcopyrite.

Exploration took place over a length of more than 1.6 kilometers, to a depth of 570 meters in 19 levels of galleries, with an average spacing of 30 meters. The mine had 6 shafts, the main one being the Sta. Bárbara shaft. Conventional underground mining was used until 1973, when in situ leaching with sulfuric acid was introduced.

A total of 4.370 tonnes of uranium oxide was produced in Portugal, of which 25% was mined at Urgeiriça and 75% from the surrounding mines located in the central region of Portugal, which were also processed at the Urgeiriça mill.

Environmental remediation of Urgeiriça legacy mine site

Mining operations from these past activities left environmental impacts caused by two tailings dams, waste rock piles, sludge deposits and various mining facilities, a uranium mill and related buildings and equipment in the industrial area, as well as risks due to the physical and safety stability of some buildings. Radiological and chemical contamination in the surrounding area, including contamination of surface water and groundwater, air, soil and sediments, had resulted in an existing exposure situation, mainly due to external gamma radiation, indoor and soil exhalation of radon gas and inhalation of dust.

Environmental remediation of the Urgeiriça mining area began in 2001 and comprises a phased, stepwise approach, as defined in the Master Plan of the Mining area of Urgeiriça, that aimed to remediate the risks to health and the environment from past mining activities. The implementation was concluded in 2022, with a total investment of more than 33 million euros. Funding was provided by EDM, mining royalties from current mining operators in Portugal, and the European Commission Cohesion Funds.

Urgeiriça Mine Water Treatment Plant details

Mine Water Treatment Plant (MWTP) new facilities were constructed in 2016, as part of the final phase of the environmental remediation works at the Urgeiriça mine (Fig. 1). The treatment system is divided into two lines, active treatment and passive treatment carried out through sedimentation and aeration tanks and phytoremediation ponds. Division criteria results from a balance between the installed treatment capacity and operation costs. In average 80% of flow rate is directed to the passive treatment and 20% to the active treatment. Mine effluents consists in subsurface outflows from two sealed tailings dams, which are collected via underground drains and directed to a leached well, from where they are pumped for treatment (Diamantino *et al*. 2023). Mine water treatment was complemented with a passive system in order to evaluate a gradual transition to a more sustainable solution in the long term, if adequate contaminant removal could be achieved without the need for continuous operation, reagent addition and energy consumption, and benefiting from a natural attenuation process in terms of effluent quality.

Active treatment line includes processes for treatment and separation between the liquid and the solid phases. Treatment of the liquid phase (Fig. 2) includes pH neutralization by adding calcium hydroxide ($CaOH_2$) to increase the pH and barium chloride ($BaCl_2$) to promote precipitation of Ra-226 and uranium, followed by decantation for solid-liquid separation, with monitoring of the effluent quality at the end of the treatment line. Two reactors for pH correction and two reactors for the addition and mixing of barium chloride are installed inside the MWTP building.

MWTP generates sludges that require additional thickening and dewatering operations prior to disposal. After chemical treatment, effluents are directed to a decantation system consisting of two circular decanters. The sludge accumulated at the bottom of the clarifier is directed to the sludge pumping well and pumped to a circular gravity thickener silo. Solid phase treatment process involves gravitational thickening of the sludges in a silo, followed by dewatering with a filter press and the use of filter bags for storage. Effluent from the thickening and dewatering of the sludge is recirculated. In order to improve the efficiency and speed

Figure 1 *Mine water treatment station at Urgeiriça mine site (Diamantino* et al. *2023).*

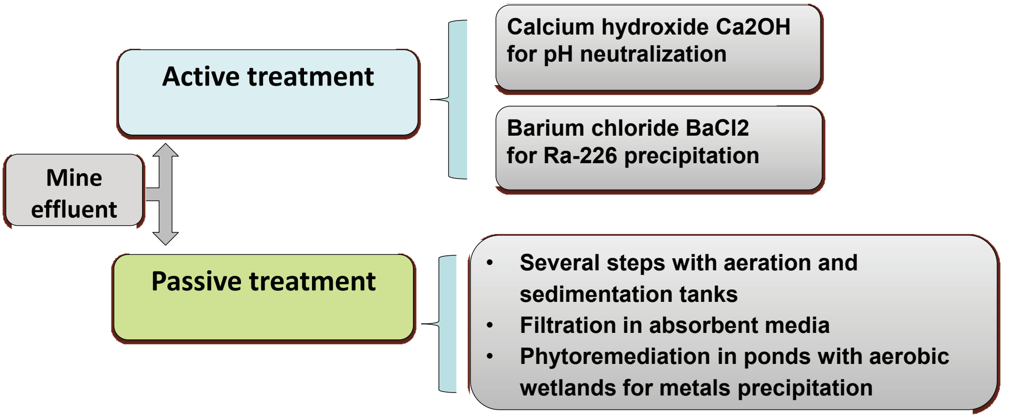


***Figure 2** Mine water treatment scheme at Urgeiriça mine site (Diamantino* et al. *2023).*

of the sludge dewatering process, polymer dosing is used on the sludge to be dewatered.

Passive treatment system

The passive treatment system essentially corresponds to a water treatment structure using principles of aeration, sedimentation, neutralization, and filtration in adsorbent media and phytoremediation. It consists of several interconnected tanks with gravitational effluent circulation.

The effluent is pumped from the leachate well through aeration cascades to primary and secondary sedimentation tanks. Aeration promotes the precipitation of iron oxides as well as other contaminants and metals. The effluent is them discharged into to the neutralization and filtration line, which consists of different filling materials, such as limestone, barite, and activated carbon, placed in big bags inside the tanks, protected at the top and bottom by limestone gravel. The total hydraulic retention time in the neutralization and filtration process, taking into account the average annual flow rate of 30 m^3/h, is approximately 126 hours.

Then the effluent from passive treatment is mixed with the effluent from active treatment before being pumped to the two phytoremediation ponds to improve the quality of the treated water. Biological treatment ponds consist of floating strips of macrophytes planted with *Juncus effusus*, *Iris pseudacorus*, and *Typha latifolia*. This solution has the advantage of providing phytoremediation of the water through a passive, natural, low-maintenance, and very simple to operate process, i.e. a phytoextraction process that consists of extracting the contaminant elements from the effluent and accumulating them in the biomass produced by the installed vegetation.

At the end of the last pond, the effluent passes through a monitoring station for continuous water quality and flow control before being released into the watercourse. If the water quality does not allow for discharge into the watercourse, the effluent can be recirculated through an existing pumping station to the beginning of the active treatment. The system was designed for an average influent flow rate of 30 m^3/hour and a maximum flow rate of 60 m^3/hour.

Chemical and radiological composition of mine effluent

Water quality control programme started in 2001. The effluent to be treated is generally acidic (with an average pH of 3.00 and electrical conductivity of 1313 µS/cm for 2018–2024), with high concentrations of metals such as manganese and iron and radionuclides. Table 1 shows the annual average concentration of chemical and radiological elements and in situ parameters, in 2023, for the effluent before and after treatment, with sampling frequencies of monthly and every two weeks, respectively.

Fig. 3 shows the annual variation of average concentrations in mine effluent, before and after treatment over the last 7 years, for sulfate, manganese, pH and total

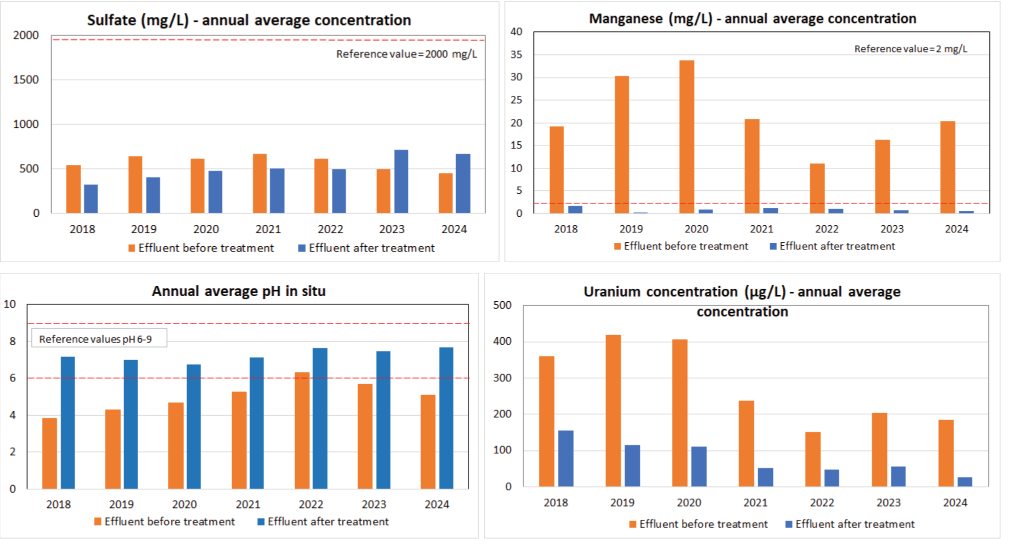


Figure 3 Annual variation of average concentrations in mine effluent before and after treatment. Dashed lines in red colour are the respective parameter reference values from permit licence for treated mine effluent discharge.

uranium concentration, with a decreasing tendency for uranium concentration and a smaller increasing tendency for pH values. Long historical data series can document this natural attenuation process after environmental remediation (Diamantino et at. 2023).

Removal efficiency rates and compliance with regulatory limits for mine effluent water discharge

The removal efficiency rates of mine water treatment, calculated on the basis of available laboratory results of chemical and radiological analysis, for the main contaminants and considering average annual concentrations in 2023, are the following: 100% for iron, 95% for manganese, 73% for total Uranium and 75% for Ra-226 (Table 1). With regard to the reference limit values for the discharge of treated mine effluent, a license permit has been issued by the national Regulatory Body for the management and protection of water resources and radiological protection (Environmental Protection Agency, Agência Portuguesa do Ambiente – APA). Table 1 shows these reference limit values defined for pH, sulfate, iron, manganese, TSS, radionuclides and Indicative Dose calculation. For radiological parameters, compliance is established for water for human consumption, taking into account the contamination history of the site. The results of this control and monitoring programme are reported annually to the Regulatory Body for compliance evaluation.

Quantities of chemical reagents and operation and maintenance costs for mine water treatment

Chemical reagents used in the active treatment are calcium hydroxide ($CaOH_2$) and barium chloride ($BaCl_2$). Fig. 4a shows the monthly evolution of the quantities of reagents used in 2023. In total, 58 m^3 of $CaOH_2$ and 2400 kg of $BaCl_2$ are used annually. As expected, higher amounts are used during the winter season when precipitation and discharge rates increase. Fig. 4b shows the monthly evolution of discharge rate and total precipitation in 2023. October was the wettest month, but this was not reflected in the increase of treated flow until the following month. The average flow rate was 21,5 m^3/h in 2023.

The operation costs in Urgeiriça MWTP, considering only the consumption of chemical reagents are shown in Table 2 and are around 16.800€/year (1400€/month). Other costs include technical staff, maintenance and cleaning operations and management of sludge produced, estimated at 14.000€/month, but shared and optimized

Table 1 Mine effluent composition before and after treatment, annual average values in 2023. Calculated efficient rate for contaminants removal. Reference values from permit licence for treated mine effluent discharge.

Parameter	Mine effluent before treatment	Mine effluent after treatment	Efficient rate	Effluent reference limits
N.º samples	12	24	–	–
pH	5.68	7.48	24%	5–9
EC (µS/cm)	1361	1086	20%	
Redox potential (mV)	179	137	–	
Temperature (ºC)	16.88	18.31	–	
Sulfate (mg/L)	715	498	30%	<2000
Chloride (mg/L)	33.20	34.99	–	
Sodium (mg/L)	42.40	35.68	–	
Calcium (mg/L)	201	181	10%	
Iron (mg/L)	11.83	0.05	100%	<2
Manganese (mg/L)	16.23	0.78	95%	<2
Total Suspended Solids TSS (mg/L)	31.6	3.95	87%	<60
Uranium total (µg/L)	203.8±30	55.8±8.0	73%	–
U-238 (Bq/L)	2.518±0.39	0.689±0.01	73%	3.0
U-234 (Bq/L)	2.213±0.35	0.593±0.01	73%	2.8
Ra-226 (Bq/L)	0.062±0.02	0.015±0.01	75%	0.5
Pb-210 (Bq/L)	–	0.05±0.02(b)	–	0,1
Indicative Dose (mSv) [(a)]	<0.10 conformity for water consumption – 15 in 20 analyses >0.10 conformity for water consumption – 5 in 20 analysis			<0.10

(a) Indicative Dose is calculated accordingly with Decree Law nº 69/2023, 21 August, from the measured radionuclide concentrations and dose coefficients set out in Table A of Annex iii to Council Directive 96/29/EURATOM of 13 May 1996. If [Σ Ci(obs)/Ci(der) ≤1] them Indicative Dose <0.10 mSv. Considering the sum of main radionuclides: U-238, U-234, Ra-226, Po-210. (b) average concentration considering 20 samples.

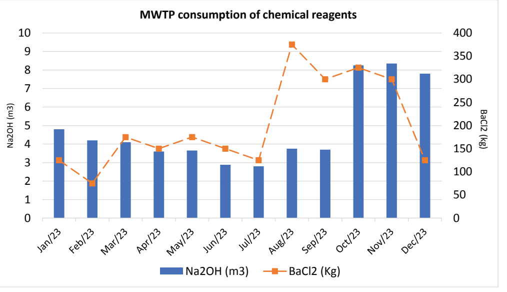
between other smaller mine sites, mainly with passive treatment systems. Permanent technical staff represent approximately 60% of the total operation and maintenance costs.

Conclusion

This paper presents an interesting mine effluent control and treatment plant that was implemented during the environmental remediation of the Urgeiriça legacy mine site, which combines a common and effective solution of an active system with the addition of chemical reagents and a more sustainable passive treatment system with aeration, sedimentation and phytoremediation processes. Operation and maintenance costs are high in the post-remediation phase, but the long-term reduction in concentrations of contaminants such as uranium is expected to allow a gradual transition to passive treatments. Mine water discharge limits are generally accomplished, for chemical and radiological parameters, but for radiological parameters compliance reference limits are very restrictive.

Table 2 Costs of chemical reagents consumption in 2023.

Reagents	Unitary Costs	Consumption	Total costs
$CaOH_2$	286€/m³	58 m³	16.588€
$BaCl_2$	2,77€/25kg	2400 kg	286€



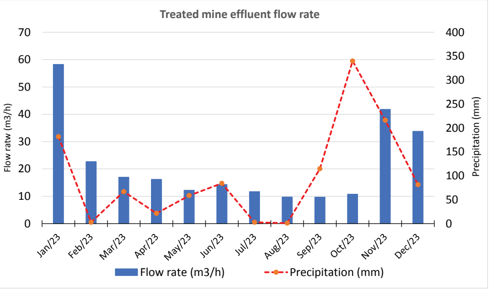


Figure 4 a) Consumption of chemical reagents in 2023. b) Treated mine effluent flow rate in 2023.

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