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Re-processing of mining waste: Combining environmental management and metal recovery?

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Abstract

Raw materials are essential for the sustainable functioning of modern societies. Access to and affordability of mineral raw materials are crucial for the sound functioning of the EU’s economy. The increasing demand in raw materials raises growing concerns regarding the mineral resources and especially metals availability. Furthermore, many metals, metalloids or rare earth elements which had no application in the past are now used for the manufacture of high added value products especially in the domain of new and green technologies. Many of them are by-products of base metals production and their reserves are very limited. Therefore, a list of strategic and critical materials was established by the European Commission in 2010. Following the rich mining history of France, there are currently some 300 mineral deposits mined or significantly explored. These are the source of large quantities of “tailings” (gangue, processing waste resulting from concentration or hydrometallurgical treatment, slag, heap).

The European Directive 2006/21/EC of 15 March 2006 on the management of waste from extractive industries oversees the permit conditions, storage, monitoring and control of the mining waste to ensure the protection of human health and the environment. The Member States have to carry out and periodically update an inventory of closed waste facilities which, cause or have the potential to cause, a serious threat to human health or the environment. Moreover, the directive encourages the recovery of extractive waste by means of recycling, reusing or reclaiming such waste. Following this, an inventory was finalized in France in 2012. 2,100 metallic and 1,300 coal tailings were identified and prioritized according to their potential impacts on ground waters, surface waters, soils and human health. Among these, 53 sites with major environmental issues were listed. These sites are and will be the subject of specific actions (environmental and sanitary studies, works of rehabilitation) in order to reduce their long-term impact both on human health and environment. While metallic elements present in the tailings are by nature non-degradable, these actions require long-term approaches.

The paper focuses on one of these sites, former Pb-Ag mines, where operations have led to the production of 200,000 m³ of mining waste. A French Geological Survey (BRGM) rehabilitation program, based on environmental studies, was designed to stop additional tailings erosion toward the river and the surrounding areas and to avoid any contacts between the contaminated materials and the population. Metals recovery could be envisaged to mitigate the remediation cost.

Indeed, old waste deposits related to past mining and metallurgical activities can be considered as significant reserves of valuable metals because the latter were not exploited or because economically recoverable metals may remain. In France, a recently started project handled by BRGM is aimed at identifying interesting old mining wastes deposits at the national level and assessing the metal recovery potential of these dumps. In a first step, it focuses on previous sites to be rehabilitated. The objective is to develop a methodology combining mining wastes environmental management and economic valorisation in a long term perspective.
1 Introduction

Raw materials are essential for the sustainable functioning of modern societies. Access to and affordability of mineral raw materials is crucial for the sound functioning of the EU's economy.

EU still has valuable deposits and much under-explored and unexplored geological potential, but their exploration and extraction face increased competition for different land uses and is required to take place in a highly regulated environment. With only 3% of the world’s ore production and 25 to 30% of the world’s metal consumption, Europe is highly dependent on imports for many raw materials. Indeed, the international context regarding mineral resources and demand is complicated by the fact that many metals, metalloids or rare earth elements which didn’t have any application in the past are now critical elements for the manufacture of high added value products especially in the domain of information, communication and green technologies (cf. Figure 1). For example, new low carbon technologies like wind turbines, electric cars, energy saving light bulbs, fuel cells and catalytic converters, require rare and precious metals for their production. Many of them are by-products of base metals production (cf. Figure 2).

These materials are increasingly affected by growing demand pressure from emerging economies and by an increasing number of national policy measures that disrupt the normal operation of global markets.

Figure 1  Evolution of metal and element use along time in industry

Following this, the European Raw Materials Initiative was published in November 2008 as an integrated strategy to (1) secure sustainable access from outside Europe, (2) improve framework conditions for extracting minerals within Europe, and (3) promote the recycling and resource efficiency of such materials. A list of 14 critical materials was established by the European Commission in 2010 (Antimony – Sb, Beryllium – Be, Cobalt –Co, Fluorspar, Gallium -Ga, Germanium –Ge, Graphite, Indium – In, Magnesium - Mg, Niobium – Nb, Platinum Group Metals - PGMs, Rare Earths - RE, Tantalum- Ta, Tungsten –W) according to the following criteria:

- Economic importance
- Supply risk (Stability/Instability and level of concentration of producing countries, Substitutability, Recycling, Aggregation)
- Environmental country risk
Figure 2  Relationship between the main “carrier” metals and the associated co-product metals (from Reuter et al. 2005). The ‘Metals circle’ shows the occurrence of minor and critical metals with the primary carrier metal ores

Taking into account the specificities of the French industrial context, silver and titanium have been added to the European list. It is important to note that in many cases a significant proportion of primary global resources of the critical metals are hosted in poly-metallic deposits. That is, these critical metals commonly do not occur on their own but along with other metals. In addition, the critical metals are usually not the major commodity occurring in the deposits and are considered as by-products of the main carrier metals. For example poly-metallic copper, nickel, zinc and lead resources will likely be accompanied by several of the critical metals.

Given the evolution of the context regarding mineral resources and the necessity to recycle, old waste deposits related to past mining and metallurgical activities can be considered as significant reserves of valuable metals because these latter were not exploited or because economically recoverable metals may remain.
2 Mining Waste context

2.1 Definition

Mining waste can be defined as a part of the materials that result from the exploration, mining and processing of substances governed by legislation on mines and quarries. The term “Mining wastes” regroups in reality different products resulting from the extractive industry. Waste rocks consist of non-mineralized and low-grade mineralized rock removed from, around, or within the orebody during extraction activities. The cut-off grade that differentiates low-grade waste rock from useable ore is an economic distinction and may vary over time. Processing waste or tailings are the waste solids or slurries that remain after the treatment of minerals by separation processes, e.g. crushing, grinding, size-sorting, flotation and other physico-chemical techniques, to remove the valuable minerals from the less valuable rock. More specifically, slags are residues from the smelting pyrometallurgical process.

They have to be differentiated from the overburden which corresponds to the surface material (topsoil, rock) removed during surface mining operations to expose the ore beneath.

2.2 Quantities in Europe and France

According to Eurostat estimations (Eurostat, 2010), mining and quarrying wastes account for more than 720 million tonnes in 2010, corresponding to 28% of the total waste production in Europe (27 countries), and 1 million tonnes in France.

It is difficult to obtain precise numbers concerning the historical mining wastes. Except for the Promine Databases, to be discussed, no Europe-wide database exists to date on the location and the physical and chemical characteristics of mining waste and other industrial deposits. Following the rich mining history of France, there are currently more than 300 mineral deposits mined or significantly explored. These are the source of large quantities of mining wastes. According to a study carried out by the French Geological Survey (BRGM, 1997), it was estimated that 130 million tonnes of non-ferrous mining wastes exist in France. And according to a European study carried out by BRGM (BRGM, 2001) 1.2 billion tonnes of tailings (see definition below) were estimated in Europe (12 countries) with 78 million tonnes in France. These numbers are to be put in perspective as they were obtained with questionnaires and based on ore production.

More recently, as a result of the European Directive inventory finalized in France in 2012, 2100 metallic and 1300 coal tailings were identified and prioritized according to their potential impact on ground waters, surface waters, soils and human health. Among these, 53 sites with major environmental issues were listed. These sites are and will be the subject of specific actions (environmental and sanitary studies, works of rehabilitation) in order to reduce their long-term impact both on human health and environment. Metallic elements present in the tailings being by nature non-degradable, these actions require long-term approaches.

2.3 Environmental impacts of mining wastes

Solid mining wastes are stored in piles or in tailings impoundments around or near mine sites. These storage facilities allow for the physical containment of the mining waste but they cannot completely prevent environmental issues. Air and soil can be contaminated by the generation and dispersal of dust, and groundwater and surface water can be contaminated by seepage through embankments or through the base of the tailings pile. In summary, these wastes can affect the environment through one or more of the following intrinsic criteria:

- Chemical and mineralogical composition,
- Physical properties,
- Volume and surface occupied,
• Waste disposal method.

They typically contain large fractions of sulphide material which are vulnerable to alteration, as they come into contact with air and meteoric water. In the absence of sufficient neutralizing minerals such as calcium carbonates (Juillot et al., 1999), acid mine drainage (AMD), also called acid rock drainage (ARD), is generated. A worldwide estimate assumes that about 20,000 km of river and 70,000 ha of lake and reservoir areas are seriously damaged by acidic mine effluent (Johnson and Hallberg, 2005). The control and mitigation of AMD mining wastes alone is considered to be one of the major environmental challenges facing the mining industry worldwide (International Network for Acid Prevention 2011). This is evidenced by sites such as the Rio Tinto estuary in Spain, where surface water contamination is still present from historic mining as early as 4500 years ago (Leblanc et al., 2000).

Most dramatically, tailings dams can fail. Since 1970, over 70 major failures of impoundments have occurred around the world. Many of these have resulted in short- and long-term damage to ecosystems, in significant impact on communities and most seriously, in the loss of over 1000 lives (Blight, 2010; Fourie, 2009). The Aznalcóllar–Los Frailes dam failure in 1998 (Grimalt et al. 1999) is unfortunately well known and led to the discharge of 4.5 million cubic meters of toxic tailings slurries into surface water (Achterberg et al. 1999); the rehabilitation has cost approximately €250 million.

2.4 Regulation context

The 2006/21/EC European Directive on the management of waste from extractive industries oversees the permit conditions, the storage, the monitoring and the control of mining waste. The purpose is to ensure the protection of human health and the environment. The member states have the obligation to carry it out by 2012 and periodically update an inventory of closed waste facilities. The Directive encourages the recovery of extractive waste by means of recycling, reusing or reclaiming such waste.

Moreover, in January the 28th 2012 the Consultative Commission on Industrial Change (CCMI) on the processing and exploitation of industrial and mining waste deposits in the European Union made several observations and proposals:

• no Europe-wide database exists to date on the location and the physical and chemical characteristics of mining waste and other industrial deposits,

• it suggests to reassess the economic potential of waste resulting from old industrial activities and to make this domain environmentally sound, and

• it suggests stimulating research and innovation on technology for reworking and rehabilitating old overburdened waste repositories. The European Innovation Partnership on Raw Materials could provide a vehicle to stimulate research in this area and possibly provide funding for pilot projects.

Providing long term access to the available mineral resources therefore requires more focus on sustainable mining, both on research for environmentally sound mining and processing technology, as well as on the social and economic aspects of mining. To counteract the steady increase in global demand for primary mineral resources and to reduce the negative social and environmental impacts associated with meeting this demand, it is necessary to increase material efficiency in manufacturing processes, to search for new substitute raw materials and to recycle materials more widely and more effectively, through technological innovations. Reprocessing of mining wastes, prioritizing those with acute environmental impacts, is a way to improve this metal recovery while protecting human being and the environment.

The following sections present an example of a mining waste rehabilitation project.
3 From a classic rehabilitation toward a global environmental management

3.1 Site presentation

In Auvergne, vein deposits of argentiferous galena, sphalerite and pyrite with arsenic were mined so that Pontgibaud was one of the largest mining and metallurgical districts in Europe for lead in the Nineteenth century. 50,000 tonnes of lead and 100 tonnes of silver were produced up to 1905 when the metallurgy ended.

The legacy of the mining activity consists of:

- 70 km of galleries and 3,000 m of mine shaft (Gallo-Roman period and the nineteenth century);
- 30 adits and 20 minesshafts;
- 4 areas of waste rocks and tailings sites.

The mining wastes are constituted of rock wastes, tailings and slags. The total volume of mining waste is around 200,000 m$^3$, distributed as follow:

- Site 1: 78,000 m$^3$ over 23,000 m$^2$
- Site 2: 87,000 m$^3$ over 45,000 m$^2$
- Site 3: 33,000 m$^3$ over 8,800 m$^2$
- Site 4: 4,000 m$^3$ over 4,800 m$^2$

The four sites are located along the Sioule River. Since no vegetation grew on the tailings they are subject to erosion by rainfall and wind. In consequence they impact the river sediments and the surrounding areas.

3.2 The project of rehabilitation

Firstly, all the 60 adits and shafts were closed by BRGM in 2009. The operation founded by the French State was performed in order to avoid any risks such as a deliberate or accidental entry, ground instability due to the opening, the circulation of fluids (water or gas). The treatment solutions included a backfilling and closure with a self-supporting plug for the mineshaft, and for the adits closures while keeping a specially adapted opening for bats and snakes.

Secondly, an environmental study was carried out, according to a source – pathway –target methodology, to bring out the potential impacts of the 4 mining wastes sites toward human beings and the environment. The study focused on:

- source evaluation (volumes, heavy metal content, etc.);
- groundwater;
- surface water, including sediments;
- sensitivity of the surrounding environment (threatened or vulnerable species according to Natura 2000, groundwater and surface water uses, etc.)
- non-controlled reuse of tailings by local population;
- site uses.

The tailings contain high concentrations of lead (between 6 and 70 g/kg), arsenic (0.9 to 8 g/kg), cadmium (4 to 107 mg/kg), zinc (0.8 and 2.3 mg/kg) and chromium (0.01 to 0.1%) and concentrations can reach locally 8% of lead, 1.5% of zinc and 0.9% of arsenic. These elements are environmentally undesirable or dangerous to human health.
The sediments are clearly impacted. The geochemical background is estimated at respectively 90 mg/kg and 74 mg/kg for arsenic and lead. Concentrations immediately downstream of the sites are between 300 and 740 mg/kg for arsenic and between 4,000 and 11,000 mg/kg for lead. Further downstream, concentrations are between 100 and 130 mg/kg and between 400 and 1000 mg/kg respectively for As and Pb.

Impacts on Sioule River are measurable but low due to the dilution and the absence of AMD (presence of basalts and probably because a large part of the sulphides are already oxidized). The geochemical background for As and Pb is respectively of 3 µg/l and 0.5 µg/l, and the analyses show the following concentration:

- Immediately downstream: 7 µg/l for As and 16 µg/l for Pb;
- Further downstream: 5 µg/l for As and 1.5 µg/l for Pb.

Aquifer located below flows in volcanic slag and ash layers between layers of basalt and no groundwater impact was identified.

Different activities leading to a direct contact between human beings and the mining waste were identified (such as mountain biking, practice of motorcycle, etc.).

These impacts are synthetized in the following conceptual scheme focusing on site 2 (in Figure 3).

![Figure 3 Conceptual scheme of site 2 impacts](image)

Following these results, a rehabilitation program was designed to stop additional tailings erosion toward the river and the surrounding areas and to avoid any contacts between the contaminated materials and the population.

A flora and fauna study is ongoing to characterize the sites impacts according to Natura 2000 regulation. It is used to guide the rehabilitation design so that the sites residual impacts will be reduced as much as possible. The flora and fauna study recommendations include the rehabilitation works period.

The rehabilitation works will include several operations:
• Tailing and rock wastes earthworks to reduce the mining wastes footprint and to obtain final slopes reducing the rainfall infiltration while reducing erosion;
• The implementation of a 80 cm thick topsoil with amendment if necessary;
• Revegetation with local species allowing for a long term protection of the cover against erosion;
• Surface water management works (trenches, drainage).

Figure 4 illustrates the conceptual scheme of the works planned on site 2. In this case, in addition to the operation described previously, the rehabilitation program will include the restoration of former lagoons to implement a wetland. This wetland area, collecting all the waters drained from the tailing via drainage groins, will help reduce the metal load toward the river.

**Figure 4  Conceptual scheme of site 2 rehabilitation works**

The cover design is the key point of the rehabilitation program as it is the only barrier to avoid contact and spreading of the mining wastes toward the environment. The main difficulty is due to the timescale as the metal pollutants are by nature non degradable. By consequence the cover must last for the long term, and the only way to succeed involves the durability of the vegetation. The plant species will be selected according to the recommendation of the fauna and flora study and a program will be necessary to monitor the evolution of the plants distribution along time.

Studies should be carried out to investigate the influence of the organic input (organic amendment in the top soil and organic input due to development of the vegetation) on the evolution of the bioavailability of the metals present in the mining waste.

Site 2 rehabilitation works are estimated to cost 950,000€, the largest items of expenditure being the mining wastes cut and fill works and the cover implementation (20,000 m²). Considering the 95,000 m³ of mining waste (tailing and rock wastes) the unit cost of the rehabilitation is 10€ per cubic meter.

Given the volume of mining wastes all over France and Europe a global rehabilitation program seems difficult to envisage because of the cost which will be generally supported by the community. One ought to consider a prioritization in the site rehabilitations, for example based on the mining wastes national inventories requested by European Union. Given the possible presence of strategic elements in the mining waste, it is relevant to include the economic potential of their recovery in the global rehabilitation strategy.
3.3 A research project regarding metal recovery

Indeed, old waste deposits related to past mining and metallurgical activities can be considered as significant reserves of valuable metals because they were not exploited or because economically recoverable metals may remain. But site documentation is rare and applicable methodology to recover these metals is not so well defined.

The following paragraph relates a number of studies concerning mining waste reprocessing. Thereby, in the 90s bioleaching process was used to recover cobalt in Uganda from pyritic wastes previously accumulated during the exploitation of a copper mine (Morin et al., 1993; Morin, 1997). In 2005, a study (Goktepe, 2005) related the recovery of lead from lead-zinc mining wastes using a multi-gravity separator. Peek et al. (2011) focused on nickeliferous pyrrhotite tailings containing 0.8 % of nickel. Fosso-Kankeu et al. (2011) tested the biological recovery of nickel from tailing. In Germany, Kamradt et al. (2012) studied the recovery of Cu and Ag. A recently started project in central Sweden identified many mineralizations and deposits in this region, the majority of them being associated with mine dumps with high average rare earth elements contents (Högdahl et al., 2012).

In France, a recently started project VALODEM handled by the French geological survey (BRGM) is aimed at identifying interesting old mining wastes deposits at the national level and assessing the metal recovery potential of these dumps (Guézennec et al. 2013). The following methodology is under development: (1) crossing several databases and information from BRGM archives, and using selection criteria (waste quantities, metal content, waste age, metallogeny, mineralogy, process characteristics, etc.) to select wastes deposits of interest (2) testing of on-site sampling methodology completed by lab analysis (3) testing of pilot scale metal recovery technics to assess the economic potential of the sites.

Three databases were used for the cross-analyses (1) ProMine Anthropogenic Concentrations (AC) databases (Cassard, 2012; Cassard et al., 2012) (2) BDSTM (French Database of Sites and Mining Titles) and (3) Dechminue.

ProMine is an ongoing European Union (EU) project (2009-2013), focused on “Innovative concepts and processes for strategic mineral supply and for new high added value mineral-based products”. A major goal of the project was to develop a Pan-EU GIS data management and visualization system for natural and man-made mineral resources (http://ptrarc.gtk.fi/ProMine/default.aspx). The ProMine AC database focuses on major anthropogenic concentrations and includes data regarding volume or tonnage and metal content (i.e. possible presence of strategic metals). The total number of records in the database is 3,412, among which 644 are located in France (Figure 5).

The BDSTM (Bouroullec et al., 2001) Database Sites and Mining Titles is used by the BRGM to identify mining sites that require environmental monitoring. This database includes environmental data but also data from mineral deposits, location of tailings and sometimes their tonnages and grades.

The Dechminue database (Thomassin, 2001) was developed during the years 2000 and 2001. This database is derived from an European project; it pinpoints 756 mining wastes sites over 9 European countries (Bulgaria, Finland, France, Germany, Greece, Ireland, Portugal, Spain and Sweden) with their location, the period of activity, the type of process and the type of waste generated.
The querying and cross-analysis of these databases led to the selection of 83 old mining sites. A last selection step based on expert opinion allowed narrowing the list to 12 sites, corresponding to 70 million tonnes of mining waste. A further literature review was carried out on these 12 sites, including tonnages, ore residues concentration analyses, methods of forming deposits and ore processing methods that have been used in their operations.

The research program focused in a first step on site 2 described previously and this for different reasons: (1) it provides a representative volume of 87,000 m$^3$ of mining wastes with a possible presence of remaining silver especially as no flotation was performed (2) the gravimetric separation handled at that time did not enable to treat particles lower than 100 $\mu$m, which usually lead to significant losses of silver (3) the BRGM is already involved on the rehabilitation of this site.

On-site XRF chemical screening analyses were performed on surface and in depth (63 points for 44,900 m$^2$ and 52 bulk samples). They were completed by laboratory analyses. Analyses and interpretation are still ongoing but the first results demonstrate fairly consistent metal contents with median concentration of Pb, Fe, S, Ag of respectively 16,400 ppm, 24,400 ppm, 46,800 ppm and 43 ppm (Guezennc et al. 2013). The preliminary results obtained from analysis of particle size fractions demonstrate enrichment in lead and silver for particles lower to 80 $\mu$m and to a lesser extent for particles greater than 800 $\mu$m.

Figure 5 Past Mining and metallurgical sites in France (with major commodities) with mining wastes, extracted from the ProMine Anthropogenic Concentrations database (Guézennec et al., 2013)
Further analyses will be performed to design a process flowsheet for the recovery of silver (ICP-AES, X-ray diffraction and microscopic observations). The next step will be to implement a pilot to test this recovery. Other sites will be investigated using the same methodology.

In the end, the project will evaluate the economical balance of the mining waste reprocessing using a cost and benefits approach including (1) the economic value of metals recovered (2) the cost of the reprocessing (3) the cost of the remaining materials final and long term safe disposal and (4) the environmental benefit of this final disposal.

3 Conclusion and perspectives

The increase in demand (quantity and diversity) and metal prices leads to a renewed interest in the historical mining waste which can be considered as potential source of base metals and critical metals.

One major difficulty regarding mining waste reprocessing concerns the quantification and distribution of elements and minerals on all scales from the nano-scale to the macroscale in space and time (for example considering the oxidation of sulphide). Developing predictive tools and reliable, field-tested modelling of long-term mining waste behaviour are among the greatest challenges. Such data and tools are needed to establish the recycling and reuse potential of these materials; geochemists and mineralogists have a significant role to play in this endeavour.

Similarly, innovation is needed to design mobile reprocessing units designed for the recovery of metal from mining waste deposits of small to medium size while taking into account their accessibility.

Any operation will require a cost and benefit approach to ensure its feasibility. This approach should take into account restocking of the remaining mining wastes after their reprocessing:

- To ensure their long term safe disposal so that they require minimal and, if possible, ultimately no monitoring, control and management of the closed mining waste facility,
- To prevent or at least minimise any long term negative effects for example attributable to migration of airborne or aquatic pollutants from the mining waste facility. Thus, the estimated costs for total worldwide liability associated with the current and future remediation of acid drainage are approximately US$100 billion (Tremblay and Hogan 2001),
- To ensure the long-term geotechnical stability of the storage facilities.

This new approach should consider the following strategy:

- Engage the operation if the economic value of the recovered metal is superior to the reprocessing costs and the final disposal of the remaining mining wastes,
- If not, include the long term environmental and social benefits (especially regarding ground water and surface water impact reduction) of the remaining materials long term safe disposal.

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