The Abandonment of the Killingdal Sulphide Mine, Norway: a Saga of Acid Mine Drainage and Radioactive Waste Disposal.

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ABSTRACT

Killingdal mine, located in phyllites of the Central Norwegian Caledonides, has been actively worked for Cu, Zn and S for over a century. The mine was once relatively dry, the total natural groundwater inflow estimated to be of the order of a few l/min. Large volumes of mine spoil were deposited around the higher of the two access portals, the Killingdal shaft. On closure, in 1989, the leachate from the spoil tips was directed into the shaft at a rate of c.40 l/min, causing a rapid rise in water level in the mine. The rate of rise (c.7 m per month) and water chemistry (pH = 2.5, $SO_4^= = 26,000$ mg/l, Zn = 5600 mg/l, Cu = 890 mg/l, Cd = 13 mg/l) were quantified in the course of an assessment of a proposal to deposit radioactive waste in the mine. This proposal was rapidly dropped. The projected filling of the mine, and breakout of acid mine drainage from the lower, Bjørgan shaft is expected in c.40-50 years' time. Various amelioration strategies are discussed.
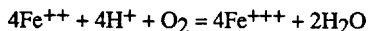
INTRODUCTION - THE GEOCHEMISTRY OF ACID MINE DRAINAGE

Mining allows the introduction of oxygen to the deep geological environment and thus causes the oxidation of minerals which are in a reduced state. The most common family of such minerals are the sulphides. Unlike most geochemical weathering processes, the oxidation of some sulphides (of the type MS_2) leads to the production, rather than the consumption, of protons (i.e. acid). The iron sulphide, pyrite, is ubiquitous in most metal
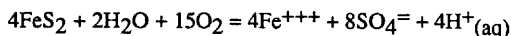
sulphide deposits.

$$2FeS_2 + 2H_2O + 7O_2 = 2Fe^{++} + 4SO_4^= + 4H^+_{(aq)}$$
Pyrite + water + oxygen = iron(II) + sulphate + acid
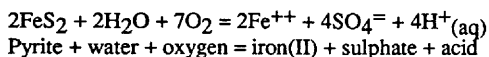
Further oxidation of iron(II) to iron (III) consumes some protons:

$$4Fe^{++} + 4H^+ + O_2 = 4Fe^{+++} + 2H_2O$$

but the overall reaction is still acid-producing:

$$4FeS_2 + 2H_2O + 15O_2 = 4Fe^{+++} + 8SO_4^= + 4H^+_{(aq)}$$

The acid released by these reactions may be partially consumed by secondary weathering or dissolution of carbonate or silicate minerals, a process which increases the pH of the water but which may release still more base cations and metals, potentially increasing the total dissolved solids of the drainage water.

Other sulphide minerals will oxidise similarly, releasing heavy metals and sulphate. It is interesting to note, however, that common sulphides of the form MS do not release acid, e.g. sphalerite:

$$ZnS + 2O_2 = Zn^{++} + SO_4^=$$
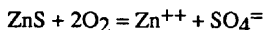
Dissolved iron (and other heavy metals) is typically soluble under the reducing and acidic conditions prevalent in the mine, but may undergo oxidation, hydrolysis and/or precipitation as oxyhydroxides (or ocre) under higher redox or pH conditions, for examples,

$$Fe^{+++} + 3OH^- = Fe(OH)_3$$

Other heavy metals will coprecipitate out with iron oxyhydroxide or be adsorbed onto the iron oxyhydroxide mass. Such ochre deposits can result in discoloration of the beds of recipient watercourses and may also damage the aquatic ecosystem, particularly as regards benthic flora and fauna.

All of the above reactions, from pyrite oxidation to ochre deposition, are bacterially mediated (Banks 1992).

THE CALEDONIAN SULPHIDE ORE PROVINCE

The Caledonian mountain chain and, in particular, the base metal mineralisation province of Trøndelag (Mid-Norway) hosts Norway's richest strata-bound sulphide resources of iron, copper, sulphur and zinc, and also some of Europe's worst mine-drainage related pollution (Iversen & Johanessen 1984). Some of Norway's finest salmon fishing rivers are found in the region; a source of considerable conflict with mining activities. The Orkla, Gaula, Orva, Hitter, upper Glomma, Stallvika, Skorovatn/Grøndal and Bleikvass Rivers have all been classified by the State Pollution Prevention Agency as being "significantly" or "highly" polluted (Grade 3 or 4) as a result of release of Cu, Zn, Fe, Pb and Cd (Schartau 1992) from natural exposures of sulphide minerals, mine drainage, gangue and slag heaps and processing works. Intensive exploitation of the mines, typically for Cu and Zn, commenced in the 17th and 18th centuries although, during more recent periods of their history, sulphur (for sulphuric acid production) has been the economic driving force for mining activities.

Killingdal Mine

The two conformable ore bodies at Killingdal (Figure 1) , the North and South Ores,

were found in 1674 and 1791 respectively, and are composed of 1.0% Cu, 10.6% Zn, 34% Fe, 42% S and 1.9% Cu, 5.9% Zn, 41% Fe and 48% S respectively (Rui 1973). They are composed dominantly of pyrite with subsidiary sphalerite and chalcopyrite and minor pyrrhotite, galena, arsenopyrite, tetrahedrite, bournonite, mackinawite and molybdenite. The ores are emplaced in the vicinity of the boundary between the calcareous Kjurudal phyllite Formation and the Hersjø Formation, consisting dominantly of hornblende metabasalt schists. Hydrothermal metasomatism related has led to
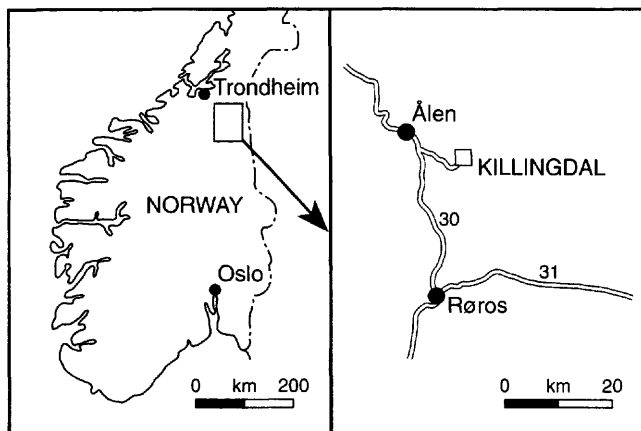

Figure 1 Map of Southern Norway, showing location of Killingdal Mine
(after Henriksen et al., 1992)

successive quartz-muscovite and chlorite alteration envelopes in the wall rocks adjacent to the ore body. The genesis and mineralogy of the ore are described by Rui (1973). Working was only sporadic prior to 1850, but by 1984, 3 million tonnes of ore had been extracted. The mine was closed in the late 1980s. The mine is over 1 km deep, and has a relatively simple structure consisting of two shafts, the Killingdal shaft (along the ore bodies) and the Bjørgan Shaft (a transport shaft at right angles to the ore). These shafts meet at the so-called Central Station level at -254 m below sea level.

During the working of the mine, spoil was mainly deposited on the upland region near the head of the Killingdal shaft. Prior to the 1980s, highly contaminated run-off from these spoil heaps was allowed to flow into streams draining into the Gaula River. Yearly fluxes of 3.7 tonnes Cu, 25 tonnes Zn, 79 tonnes Fe and 238 tonnes S were estimated by Iversen and Johannessen (1984) to be derived from these wastes, accounting for 58 %, 99%, 73% and 63% respectively of the fluxes of these elements in the heavily contaminated Gaula. Following closure of the mine in 1989, the spoil heaps were covered by low-permeability till and a drainage system was installed. Drainage water was collected and diverted into the Killingdal Shaft. The prevailing reason for adopting this policy seems to have been "out of sight, out of mind", the mine being regarded as an inexhaustible receptor of mine waste leachate.

MINE HYDROGEOLOGY

The mine has always been naturally relatively dry, being constructed in low-permeability phyllites, and no deep pumping was necessary during working. Only 2-3 l/min of drainage water ran past the Central Station level, only to be evaporated away by the time it
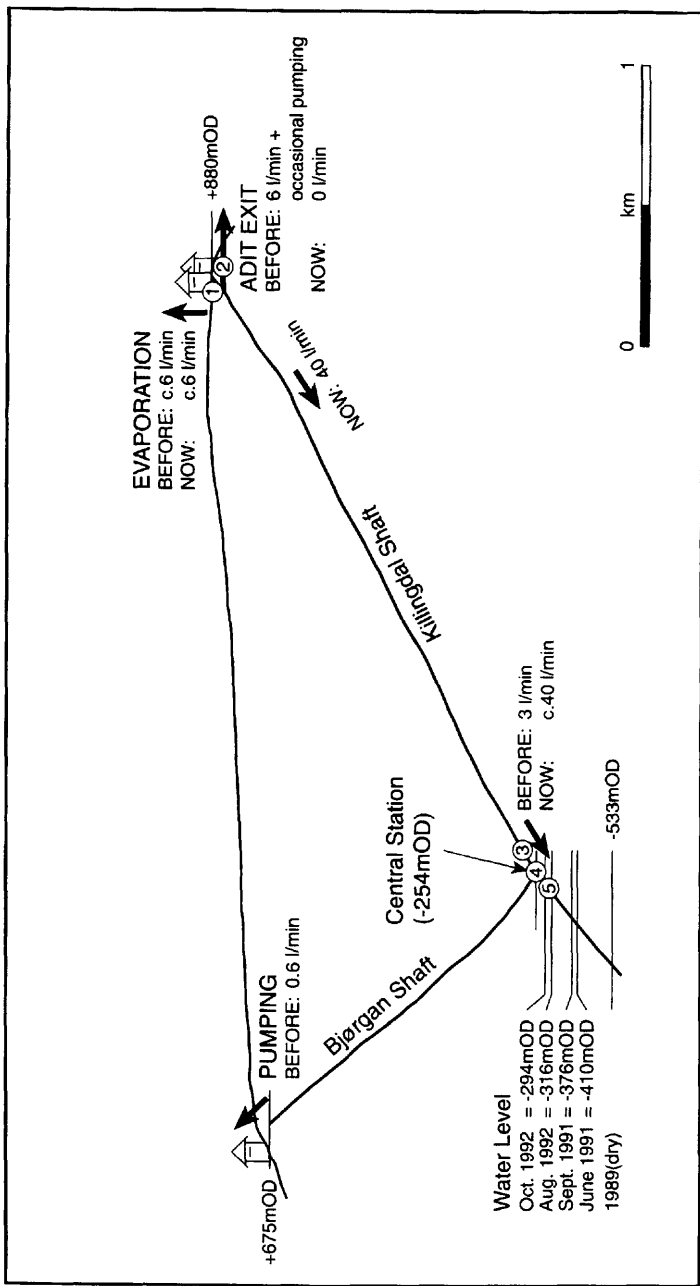
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Fig 2. Cross section through Killingdal Mine, showing water balance before and after remedial works at spoil tips and the rising water level in the mine (after Banks 1993). mOD = metres over sea level. The locations 1-5 illustrate the sampling points corresponding to the analyses in Table 1.
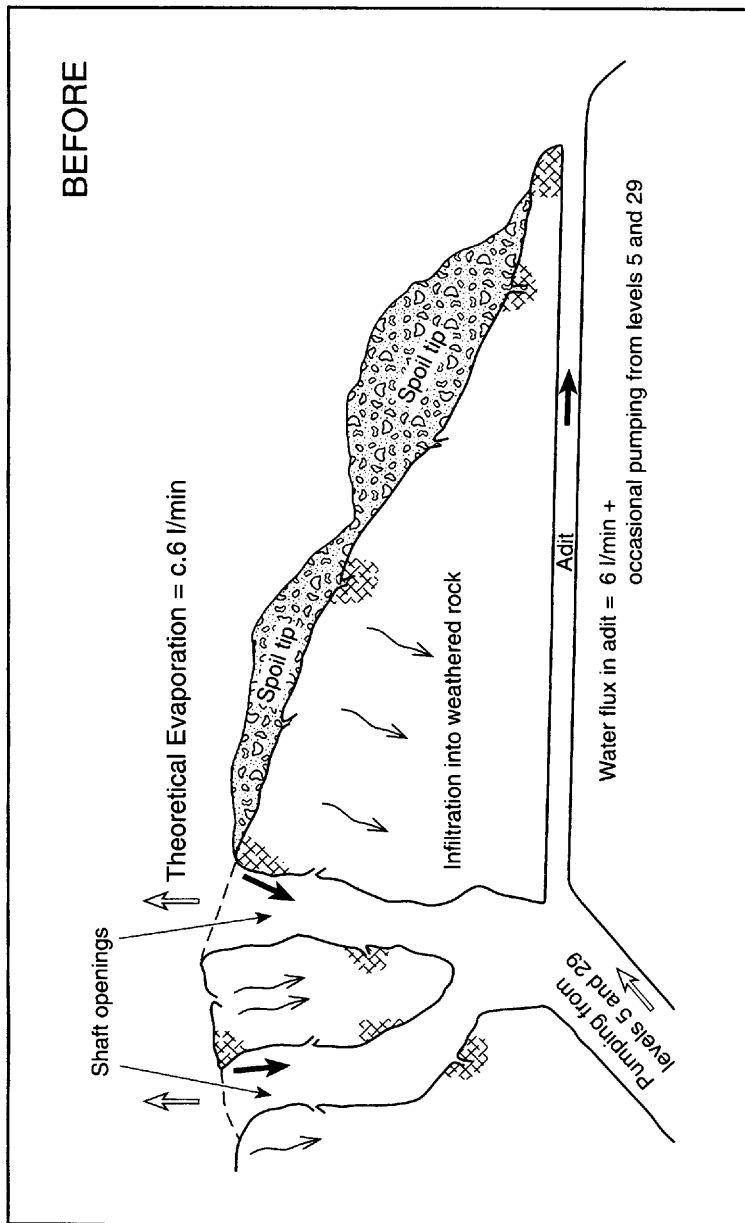
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Fig 3a. Cross section through head of Killingdal Shaft, showing water balance before remedial works at spoil tips (after NOTEBY 1992).
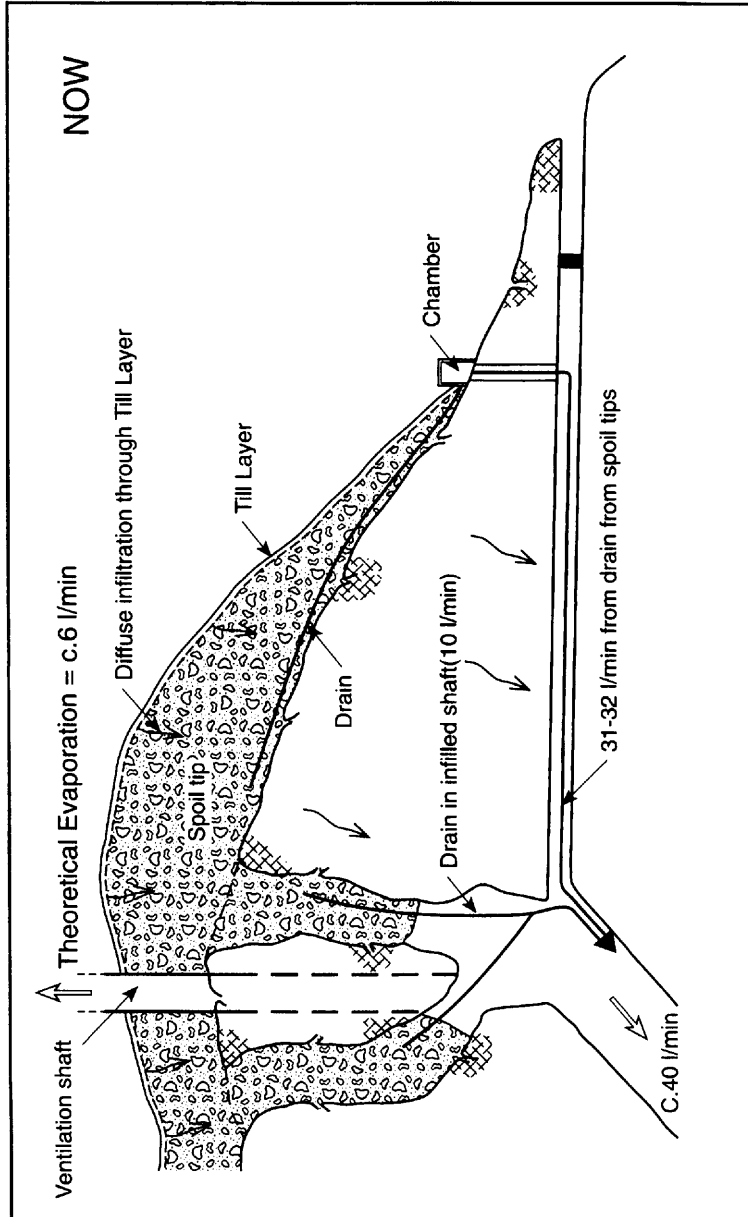
Fig 3b. Cross section through head of Killingdal Shaft, showing water balance after remedial works at spoil tips (after NOTEBY 1992).

reached the base of the mine. Other water fluxes removed from the mine comprised (Figures. 2-3):

- °   0.6 l/min water pumped from shallow levels of the Bjørgan Shaft,

- °   6 l/min drained from the horizontal adit near the top of Killingdal shaft,

- °   variable, but relatively small quantities, pumped from intermediate levels in the Killingdal shaft on an *ad hoc*, as needed basis

In addition, up to 6 l/min (estimated) may have been removed by evaporation. The high evaporation rate is due to elevated temperatures at depth (exothermal pyrite oxidation and geothermal gradient) and extremely powerful air currents through the mine (differential barometric pressure and temperature between the two mine portals). The total seepage of natural groundwater to the mine complex (excluding the adit) is unknown but is likely to have been of the order of a few l/min.

This hydrogeological situation changed following 1989 when leachate from the mine waste tips was diverted down the Killingdal shaft at a rate of 40 l/min.

RADIOACTIVE WASTE DISPOSAL AND HYDROGEOCHEMISTRY

The rate at which the mine was filling up with this leachate was only uncovered when the Norwegian Government authorities decided that a nice, deep and reportedly dry mine like Killingdal would be ideal for disposal of Norway's modest amounts of low and intermediate level radioactive waste ("out of sight, out of mind", again ?). The environmental assessment group appointed for this proposal visited the mine and was able to report (Henriksen et al. 1992, Banks 1993) that the mine was filling rapidly with water. This mine water was of highly environmentally aggressive quality; at its worst, at depth in the mine, it had the following parameters:

- °   pH 2.5
- °   26,000 mg/l $SO_4^=$
- °   an impressive array of heavy metals (dissolved Zn, Fe, Cu and Cd up to 5600, 3700, 890 and 13 mg/l respectively)

The rate of water level rise was found to be some 7 m per month (Noteby 1992), with the possibility of some seasonal fluctuation. The quality of water was seen to decline during its path from the surface, down the Killingdal shaft, to the standing water level (Banks 1993), due to either (or both) (Table 1, Figure 4):

- °   further oxidation and dissolution of ore minerals in the mine or
- °   upconcentration due to evaporation.

The fact that Na and Cl concentrations remain at constant, low levels (Table 1) indicates that evaporation is not likely to be the cause of the increase. It is worth noting that Ca and Mg concentrations increase dramatically in the inflowing spoil leachate with depth, possibly indicating the dissolution of calcite and other Ca/Mg minerals from the rather calcareous host rocks. This may exert some buffering effect on pH and partially explain why pH remains relatively constant (although very low) with increasing depth.

The rate of filling was expected to be relatively constant at 40 l/min or 21,000 m³/yr, given the fact that the water is dominantly derived at a constant rate from an external surface source (rather than by groundwater seepage, which would decrease as the water level increased). Assuming a relatively constant rate of infill and a total worked volume of some 1,000,000 m³, overflow at a rate of some 40 l/min from the Bjørgan Shaft can be predicted within at most c. 40-50 years. The fact that ventilation will cease as the shaft junction (Central Station) becomes submerged, may serve to decrease evaporative losses and hasten the rate of filling.
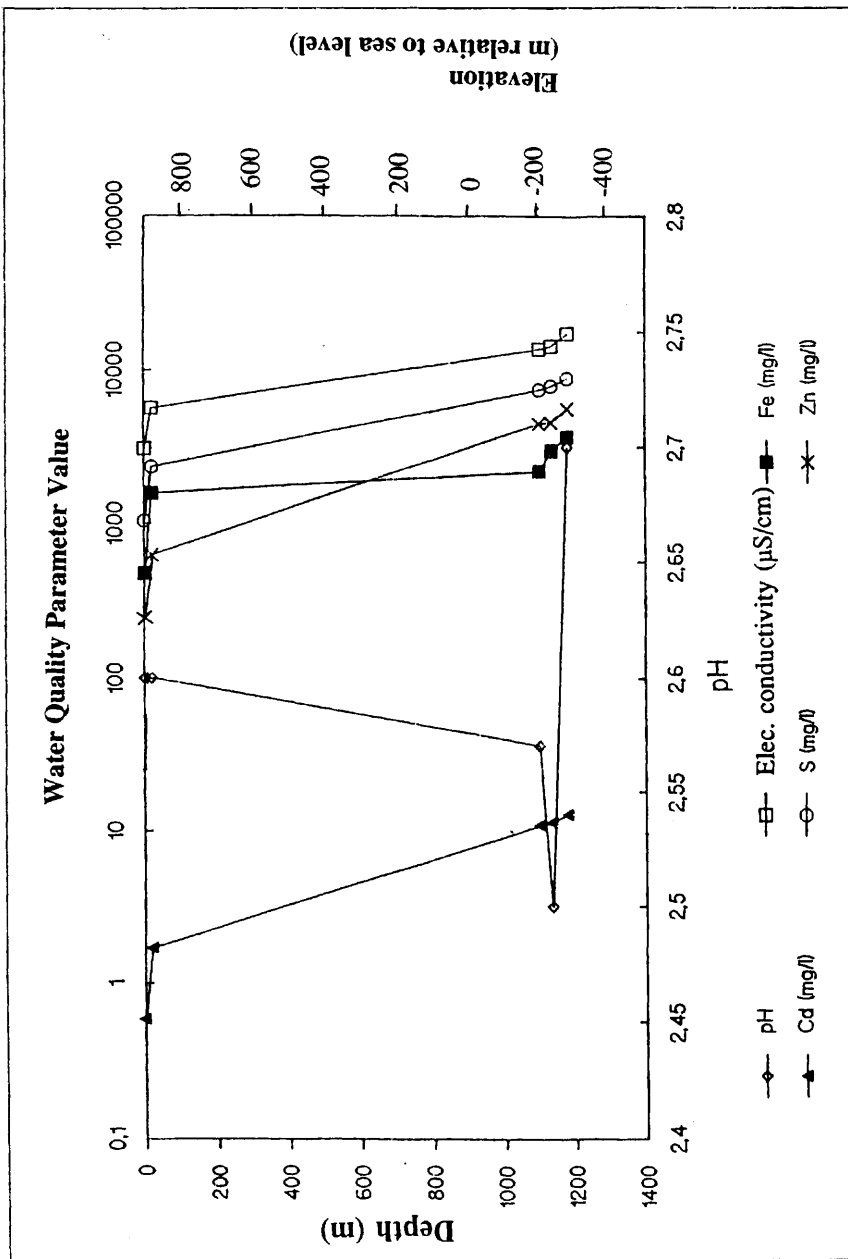
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Fig. 4. Change in chemical composition (pH, electrical conductivity, SO₄ –S, Fe, Zn, Cd) of mine spoil leachate during its passage through Killingdal mine (after Banks 1993). Data derived from Table 1 (points 1 - 5, as shown on Fig. 2).

Table 1 Variation in mine water chemistry with increasing depth. The deepest sample is from the standing water at the base of the mine, Autumn 1992 (Noteby 1992)

	Chamber (Spoil tip)	Adit	Lagoon	Central Station	Base of Mine (Standing Water)
Location (Fig. 2)	1	2	3	4	5
pH	2.6	2.6	2.6	2.5	2.7
Elec.cond. (µS/cm)	3060	5650	13700	14300	17400
Ca (mg/l)	78	143	498	547	553
Mg (mg/l)	80	180	700	782	985
Na (mg/l)	3.0	2.8	2.5	2.0	9.6
SO_4= (mg/l)	3120	6990	22400	23600	26500
NO_3-N (mg/l)	0.50	0.56	< 11	< 0.05	0.68
Cl⁻ (mg/l)	< 10	< 10	< 50	< 10	< 10
Al (mg/l)	134	254	812	778	885
Si (mg/l)	25	36	90	102	98
Fe (mg/l)	474	1570	2100	3000	3680
Cu (mg/l)	31	75	740	886	574
Mn (mg/l)	13	23	67	49	64
Zn (mg/l)	252	622	4500	4600	5640
Pb (mg/l)	< 1	< 1	0.7	< 1	< 1
Cd (mg/l)	0.59	1.7	10.6	11.4	12.8
As (mg/l)	0.083	0.37		< 0.0001	0.28
Cr (mg/l)			0.7		
Co (mg/l)			3.9		
Ni (mg/l)			1.9		

The plans for radioactive waste disposal appear to have been dropped (Henriksen et al. 1992), on a multitude of grounds: geochemical, hydrogeological, logistical and economic, the most important of which being:

º   rapid rise and projected overflow of water

º   the highly aggressive chemical nature of the mine water towards the waste and any cement/concrete encapsulation.

º   the possibility of galvanic activity and enhanced metal corrosion in the mine, due to the conductive ore body forming a significant redox couple

º   large transport distances, costs and possibly significant safety hazards associated with these.

The question of how to tackle the mine-drainage problem remains to be solved within the next few decades.

POTENTIAL REMEDIATION TECHNIQUES

Several options exist to ameliorate the effects of acid mine drainage from disused workings and waste tips.

1. Remove Mine Waste Tips to a More Suitable Place of Disposal.

This technique has been tried at the Norwegian Skorovas mine (Kopperstad 1992), where waste tips were removed to an underwater landfill in a lake, with a certain degree of success. The sheer volume of waste at Killingdal and the lack of a suitable recipient would seem to preclude such a solution. The ideal solution might have been to deposit the spoil within the mined void, seal the Bjørgan shaft and allow the mine to refill. The works carried out around the Killingdal shaft would now appear to render such an option logistically impractical.

2. Isolate waste tips.

In-situ isolation of waste tips by use of, for example, low-permeability membranes will significantly reduce the quantity of rainfall infiltration and thus the quantity of leachate generated. This was attempted to some extent at Killingdal by the use of a covering of low permeability tills, but enjoyed limited success. Experiences at the nearby Kjøli mine (Østmoe 1991) indicate that use of geomembranes can reduce the contaminant flux by over 90 % (Noteby 1992). The application of this technology at Killingdal would certainly assist in delaying and ameliorating the severity of minewater breakout. Nevertheless, such a technique would not be 100% effective and it is estimated that several l/min mine water is derived from leakage of groundwater to the mine. The mine would thus still continue to fill up and breakout of a smaller flux of contaminated water would be expected after several hundred years.

3. Pumping and Treatment of Pumped Water.

In the case of Killingdal, this is unnecessary as treatment could either be practiced at the point of entry (spoil tip area) or the eventual point of exit (Bjørgan shaft) without the need for pumping.

4. Plug Drainage Adits and Allow the Mine to Refill .

It has been suggested that the Bjørgan shaft should be plugged, allowing the mine to refill to the top of the Killingdal shaft. In order for this to work, however, several conditions must be satisfied:

- No additional old, unmapped adits should exist.

- the plug must be engineered to withstand the 200 m head difference between the Bjørgan and Killingdal shafts and be constructed of a material capable of withstanding the highly acid and sulphate rich nature of the drainage.

- the rock should be of sufficiently low permeability that mine water may not penetrate natural groundwater flow paths (e.g. fractures) to emerge as contaminated springs. The deeper in the shaft the plug is set, the better the probable engineering and hydrogeological quality of the rock, but the greater the head that the plug must withstand.

Such a solution runs the risk of catastrophic plug failure at some time in the future. It also fails to remove the source of pollution: a solution to the continued production of leachate from the waste spoil tips would have to be sought once the mine had filled up.

5. Recirculation of Leachate through Workings

Investigations at the Norwegian sulphide mine of Løkken indicate that the quality of

metal-contaminated mine waste leachate improves, as regards metal content, if it is slowly circulated through a system of sulphide mine workings (Kommunal Teknikk 1994, Lundberg 1994). There are several possible mechanisms for this:

- microbially mediated sulphate reduction and metal precipitation as sulphides
- metal exchange of toxic Cu for the less toxic Zn in the crystal lattice structure of sulphide minerals (pyrite)

At Løkken, the latter process is believed to dominate (E. Iversen, NIVA, pers.comm. 1994). Investigations are, however, underway into means by which conditions favorable to sulphate reduction could be produced. The main controlling factor appears to be the availability of an organic substrate for bacterial growth. Laboratory-scale research is currently being carried out on the use of surplus brewing yeast as "food" for bacterial communities (Lundberg 1994).

6. Treat Drainage Water

In the context of Killingdal, treatment of drainage water could take place either:

- at source (spoil heaps)
- following recirculation through the mine, at point of exit (Bjørgan shaft)

Treatment at source would have the advantage that treated water could be discharged to a surface water recipient, negating the need to inject it into the mine. Nevertheless, the mine would still fill slowly with groundwater seepage, and some form of treatment may eventually also be necessary at the Bjørgan shaft exit. Treatment at the Bjørgan shaft would have the advantage of being a single treatment location and would also benefit from natural metal exchange and sulphate reduction processes taking place within the mine (see above).

Any form of treatment would benefit from measures taken to reduce the rate of generation of leachate (i.e. isolation of spoil heaps). Treatment may take many forms from active chemical aeration, flocculation and/or precipitation followed by settlement, to biological methods. Passive biological methods have enjoyed increasing favour recently, due to their low operational costs and long-term sustainability.

The National Rivers Authority of England and Wales (NRA 1994) is currently devising pilot trials at Wheal Jane tin mine, Cornwall, of sustainable biological treatment processes, utilising arrays of rock filters, aerobic and anaerobic treatment cells, which provide ideal environments for biological processes to adsorb and precipitate metals (Bowen et al. 1994, Haddon 1994). Similar studies in the USA have demonstrated the efficacy of wetland environments at treating acid mine drainage from a range of sulphide mines in Colorado (Camp Dresser and McKee 1990).

7. Innovative Techniques

Despite the popularity of the aforementioned techniques, other ideas have occasionally been forwarded as means of prevention or treatment of acid mine drainage such as that at Killingdal:

- Application of an electrogalvanic couple to the ore body
- Application of bactericides to mines (to prevent sulphide oxidation)
- Use of heavy metal-immobilising iron bacterial bioreactors to treat water (Banks 1992)

CONCLUSION

To the best of the author's knowledge, no decision has been made regarding solutions to the problem of progressive refilling and eventual overflow of Killingdal mine, Norway, with highly environmentally aggressive acid mine drainage. Excessive haste is

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unnecessary, as breakout of mine water is not expected for another c.40 years. It is, nevertheless, the author's opinion that the optimum solution is likely to involve three components:

°   limitation of generation of leachate from spoil tips (e.g. by partial or total isolation)

°   circulation of remaining leachate through mine workings to encourage heavy metal immobilisation

°   biological treatment of waste water emerging from the Bjørgan shaft

It must, however, be recognized that other, innovative solutions may be proposed prior to eventual breakout of minewater.

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