Possibilities of remediation of neutral mine drainage – Removal and recovery of potentially hazardous elements

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Abstract: Mine water is one of the factors threatening the environment. The aim of the review article is to discuss and critically evaluate individual strategies for the remediation of neutral mine water. A critical evaluation is an essential tool to determine an appropriate remediation strategy. A wetland system is the preferred method of metal removal. However, the disadvantage is that it takes up more space compared to other methods and has a lower metal removal efficiency compared to active metal removal methods. When creating a suitable strategy, it is also necessary to assess the conditions of the mining site, which partially or completely prevent the use of the selected remediation strategy. The benefit of this review article is the processing of suitable combinations of treatment methods for the removal of potentially hazardous elements and their subsequent recovery. Future research in the field needs to focus on the analysis of the negative aspects of the environment that may disrupt or support the implementation of the selected method of remediation.

Keywords: adsorption; heavy metals; leaching; mine drainage; treatment methods

Mine drainage (MD) is an important source of environmental contamination. The most commonly mined sulfide metal ores include sulfide minerals such as galena (PbS), sphalerite (ZnS) and pyrite (FeS_2) (Novotny 1995). Acid mine drainage is the formation and movement of highly acidic water rich in heavy metals. This acidic water forms through the chemical reaction of surface water (rainwater, snowmelt, pond water) and shallow subsurface water with rocks that contain sulfur-bearing minerals, resulting in sulfuric acid. Heavy metals can be leached from the rocks that come in contact with the acid, a process that may be substantially enhanced by bacterial action (Thisani et al. 2021).

MD chemistry may vary significantly because it depends on many factors (geochemical conditions, ore deposit chemistry, host rock and tailings character). MDs are commonly characterised by a high content of dissolved toxic metals, sulfates and low pH (Braungardt et al. 2003).

MD might have a negative impact on the aquatic environment (Akcil & Koldas 2006). The MD outflows from abandoned mines into the surrounding watercourses. Water sediments act as one of the crucial heavy metal catchers. However, they are not able to fix heavy metals permanently (Yu et al. 2008). Often, streams contaminated with MD do not meet the requirements for environmental quality even after several decades. (Warrender & Pearce 2007).

Until recently, attention has been focused on acid mine drainage (AMD) (Nordstrom et al. 2000). Mine drainage does not have to be acidic in all circumstances (Banks et al. 2002). The acidity, alkalinity or toxicity depend on the specific conditions of the origin. Many mine water sources in Slovakia are characterised by a neutral pH. In general, predominated

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Table 1. Modified framework for the mine drainage (MD) classification (Thisani et al. 2020)

Class	Class description	Thresholds		
Class 0**	highly concentrated and acidic mine drainage**	pH = 0.5–3** acidity = 5–45 g/L**	total Fe = 1 000–12 000 mg/L**	SO$_4$ = 10–60 g/L** Al = 1 000–18 000 mg/L**
Class I	acid mine drainage	pH = 2.0–4.5 acidity = 0**–15 g/L	Fe^{2+} = 0**–10 000 mg/L Fe3+ = 0 mg/L	SO$_4$ = 0**–20 g/L Al = 0–2 000 mg/L
Class II	partially oxidised and/or neutralised	pH = 3.5–6.6 acidity = 0–1 g/L	Fe^{2+} = 0–500 mg/L Fe^{3+} = 0–1.000 mg/L	SO$_4$ = 500–10 000 mg/L Al = 0–20 mg/L
Class III	neutral and not oxidised	pH = 6.5–8.5 acidity = 0 mg/L	Fe^{2+} = 0–500 mg/L Fe^{3+} = 0 mg/L	SO$_4$ = 500–10 000 mg/L Al = 0–2 000 mg/L
Class IV	oxidised and neutralised/alkaline	pH = 6.5–8.5 acidity = 0 mg/L	Fe^{2+} = 0 mg/L Fe^{3+} = 0 mg/L	SO$_4$ = 500–10 000 mg/L Al = 0 mg/L
Cytotoxic metals indicator**		low = Zn 1 mg/L**	mid = zinc 25 mg/L**	high = Zn > 25 mg/L**

**indicates the revisions made to the original Hill (1968) framework

mine water sources are acid to slightly acid (pH ≤ 6) except for mine water from limestone and dolomite depositions. The neutral pH of mine effluent can be caused by:

- the absence of pyrite in the ore, thus minimising the potential for acid formation on site,
- the presence of a carbonate host rock or tailings that effectively neutralises any acidity produced (Waybrant et al. 2002).

A study (Thisani et al. 2020) on the geochemical distribution of seventy-two mining outflow sites on five continents created a modification of the classification framework of mine drainages (Table 1).

In Slovakia, there are several localities in which mine discharges occur. The most critical situation is in the Smolník area, where the discharges are characterised by low pH values (3–4) which acidify the Smolník stream (Balintova et al. 2012).

According to the annual report (Liščák et al. 2019), numerous mine outflows in Slovakia are neutral to slightly alkaline in nature, e.g., mine drainage in the Štiavnica-Hodruša mine district (Table 2).

Table 2. Characteristic values of the mine water quality indicators in the Štiavnica-Hodruša mine district in the years 2017–2019 (Liščák et al. 2019)

	Nová odvodňovacia štôlňa adit			Voznická dedičná štôlňa adit			Sludge reservoir discharge in Hodruša-Hámre			Zlatý stôl adit		
	2017	2018	2019	2017	2018	2019	2017	2018	2019	2017	2018	2019
EC (mS/m)	64	72	67	132	134	133	126	135	144	87	89	85
pH	8.32	8.16	8.09	7.49	7.37	7.57	7.70	7.37	7.32	7.44	7.36	7.30
SO$_4$ (mg/l)	242	230	205	621	618	584	590	699	643	139	141	132
NH$_4$ (mg/l)	0.03	0.03	0.03	0.04	0.05	0.03	0.11	0.27	0.31	0.03	0.03	0.03
Fe (mg/l)	0.136	0.080	0.065	1.102	0.543	0.652	0.145	0.168	1.231	0.112	0.067	0.069
Mn (mg/l)	0.109	0.041	0.021	2.200	2.140	1.975	1.136	1.900	2.410	0.483	0.428	0.367
Al (mg/l)	0.03	0.008	0.025	0.380	0.180	0.245	0.035	0.020	0.040	0.010	0.008	0.015
Zn (mg/l)	0.614	0.184	0.121	4.000	3.600	3.590	0.145	0.537	0.679	0.018	0.007	0.010
Pb (mg/l)	0.002	0.002	0.001	0.011	0.006	0.011	0.001	0.001	0.003	–	0.0003	0.0003
As (mg/l)	0.001	0.001	0.001	0.003	0.002	0.003	0.001	0.001	0.001	–	0.0003	0.0004
Cu (mg/l)	0.001	0.001	0.002	0.017	0.009	0.012	0.001	0.001	0.002	0.001	0.001	0.001
Cd (mg/l)	0.00088	0.00055	0.00033	0.0157	0.0157	0.01365	0.0002	0.0002	0.00025	0.00008	0.00005	0.00005
Ca (mg/l)	97	101	95	193	213	210	176	210	224	136	146	130
Mg (mg/l)	25	25	23	64	71	69	32	60	61	50	53	46

EC – electrical conductivity

Based on the Modified Framework (Thisani et al. 2020), mine drainages in the area can be classified as Class III Neutral and non-oxidised mine water according to the report.

In the past, quite a lot of attention was paid to remediation technologies aimed at the treatment of AMD. Abiotic (active, passive) as well as biotic (aerobic wetlands, iron oxidising bioreactors, sulfidogenic bioreactors) remediation strategies were studied in these discharges (Johnson & Hallberg 2005).

According to the report (Liščák et al. 2019), it is important to focus on remediation technologies for the treatment of neutral mine drainage (NMD) with the removal of toxic metals. By comparing Class III with Class IV of the Modification Framework, it is necessary to focus on the removal of metals (such as Al and Fe) when remediating Class III mine effluents.

Neutral mine drainage

The term neutral mine drainage is used when the mine effluent is characterised by a neutral pH and a high concentration of dissolved metals. The characterisation of seepage water and drainage water quality is needed in order to select the most appropriate remediation method. In addition, documentation of the seasonal and annual variability of the water quality is essential in setting the design criteria for the treatment system (Heikkinen et al. 2009).

To generate NMD, at least one of the following conditions must be met:

The rock contains non-acid forming sulfide minerals, e.g., ZnS, PbS or $CuFeS_2$ (Lee et al. 2018), there is enough carbonates (or other minerals) that can neutralise the acidity produced by sulfide oxidation ("Geochemical characterisation of seepage and drainage water quality from two sulphide mine tailings impoundments: Acid mine drainage versus neutral mine drainage," n.d.) after the remediation of acid-producing wastes (Maqsoud et al. 2016).

NMDs form a separate group, which is specified by an approximately neutral pH (6–8), low dissolved oxygen and medium to high concentrations of sulfates and metals, which are soluble under aerobic conditions (e.g., Cu, Zn, Cd) or insoluble (Fe, Al, Mn). Even a slightly increased iron content in the surface water can lead to the significant discoloration of a stream or river due to ochre formation (Opitz & Timms 2016).

The concentration of any element depends on the weathering of sulfidic minerals and is a function of the concentration of elements in the rock, the surface exposure of minerals (porosity, grain size, fracture density, rugged surfaces) and the susceptibility of the mineral to weathering and oxidation (climate combined with mineral properties). Amphoteric metal hydroxides (Al, Be, Cr, Fe, Tl, Sn, Zn, Cu and Ag) dissolve in water with both a low and high pH (Society for Mining, Metallurgy and Exploration 2008). Analysis of several NMD sites showed the presence of Sb at an average concentration of 175 $\mu g/dm^3$, As at an average concentration of 452 $\mu g/dm^3$ and Fe at an average concentration of 27 800 $\mu g/dm^3$ (Sekula et al. 2018).

Active and passive methods for the remediation of NMD

The selection of a treatment technology is a site-specific decision and must account for various parameters including the flow rate and water quality, and must consider economic, environmental, regulatory and social factors (Ben Ali et al. 2019). Passive systems are better known as AMD treatments (Skousen et al. 2017). The specific properties of neutral effluents do not allow the same treatment strategies to be applied as those for AMD (Tame et al. 2017). In general, for an AMD treatment, the best filter materials are those with neutralisation capabilities (e.g., lime, dolomite, fly ash, chitin and steel slag materials). The NMD characteristic implies that it is not necessary to use filter materials with neutralisation capabilities (Westholm et al. 2014). Several sorption materials have been used for NMD treatments in particular. In fact, alkaline and non-alkaline materials are often used for the removal of metals contained in NMD (Kurniawan et al. 2006; Bakatula et al. 2018). A challenging aspect is that a metal precipitation treatment in the form of (oxy)iron hydroxides cannot be applicable to these effluents due to the low Fe content (Warrender & Pearce 2007) (Table 3). In addition, in terms of the toxicity and processability, Peng and Di (1994) found that arsenic in NMD is the most problematic element due to its non-degradability (Peng & Di 1994). Carvalho et al. (2017) revealed that mine waste could contain arsenic up to 8,090 mg/kg (Carvalho et al. 2017).

When treating MD, remediation technologies are divided into active and passive methods (Viadero Jr et al. 2020). According to Trumm, the choice between an active and a passive technology is crucial for the success of the remediation technology (Trumm 2010).

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Table 3. Critical evaluation of the metal removal methods

Method of heavy metal removal	Advantages	Disadvantages	References
Precipitation	simple application applicable for most of metals suitable for large volumes of water	high reagent consumption lower removal rate may cause secondary pollution a considerable amount of sludge is formed	Ahalya and Ramachandra (2003); Yan et al. (2015); Zhu et al. (2019)
Adsorption	low operating costs applicable for wide range of metals available wide range of adsorbents simple application high removal capacity	limited by pH longer retention time stability of adsorbents during cycles when use repeatedly more difficult regeneration (some adsorbent cannot be regenerate)	Warrender et al. (2011); Sekula et al. (2018); Zhu et al. (2019)
Wetlands	good removal rate retention time can be adjusted	large space for implementation topsoil have to be removed before the next cycle higher removal rate with planted plants (plant residue need to be disposed)	Ávila et al. (2015); Sracek et al. (2018); Wang et al. (2021)
Membrane separation	high selectivity and resource recovery it is possible to obtain quality water and heavy metals concentrate	a great deal of pre-treatment is required blocking pores weak antibacterial properties limited pH range	Samaei et al. (2020)

Physico-chemical processes could be applied to both methods. The physical properties of the contaminants are used to remove the contamination in physical treatment methods (Mittal & Shukla 2019).

Active remediation methods are based on the principle of a continuous supply of the inputs to the system, which are necessary for the successful operation of the technology (Šottník et al. 2015). Active treatment systems usually require equipment (e.g., tanks, mixers, pumps), regular operation and maintenance, the continual dosing of chemicals and energy. However, they are generally considered more reliable compared to passive methods (Younger et al. 2002). Their main advantages include the efficient removal of contaminants from mine effluents (e.g., acidity, metals), precise process control so that they can be designed and operated to produce specific water chemistry, and suitability in locations where only a small land area is available (Trumm 2010).

Passive remediation methods are based on building a system that is able for some time (until its capacity is exhausted, e.g., sorption) to work without an external supply of inputs and requires only service care (cleaning the supply and discharge pipes) and monitoring (Šottník et al. 2015). Passive technologies include constructed wetlands, anoxic limestone troughs, vertical flow wetlands, open limestone canals and alkaline leaching beds (Skousen et al. 2000). Passive treatment systems for the final remediation of closed or abandoned mines should require minimal maintenance and sorption materials are usually used in a single long sorption cycle (ideally several years) before being disposed of (Richard et al. 2020a). Therefore, substrates must be stable for the same length of time and metals must be strongly bound to their surfaces (Zhou & Haynes 2010).

When treating neutral mine water with a low Fe content, it must be taken into account that due to the low Fe content, the precipitation of metals with (oxy)iron hydroxides does not help the remediation. Many of these NMDs occur in areas with a carbonate subsoil, which means that the water can be characterised by a high content of cations (Ca^{2+}, Mg^{2+}, K^{+}, Na^{+}) and carbonates, which affect the remediation by competing for sorption sites on reactive materials (Geen et al.1994). For this reason, they are considered difficult to treat by traditional passive methods (Warrender & Pearce 2007).

In the process of passive treatment, the development of media for the remediation of NMDs is still challenging, as arsenic and heavy metals should be processed simultaneously for a longer period of time and under strict conditions. Among the various materials, iron-based media have been extensively studied for the treatment of arsenic-contaminated water (Lee et al. 2018). For example, granular iron hydroxide (Driehaus et al. 2009), solid residues (Ghosh et al. 2006), hydrated iron oxide on granular activated carbon (Jang et al. 2008), granular iron medium (Selvin et al. 2002), hydrated iron oxide (Wilkie & Hering 1996) and iron hydroxide modified zeolite have been studied (Nekhunguni et al. 2017).

Methods of removing hazardous elements from neutral mining effluents

Methods of removing metals and metalloids from neutral mine effluents. Some elements, such as arsenic or chromium, are more toxic in a specific oxidation state. The main source of arsenic is weathering of arsenic bearing rock minerals and anthropogenic activities, such as mining or burning fossil fuels (Smedley & Kinniburgh 2002; Zhang et al. 2017). In water, arsenic occurs as arsenite – As(III) in oxidising conditions and arsenate – As(V) under reducing conditions (Duker et al. 2005). For arsenic removal, clay and its minerals are used with better sorption efficiency toward As(V) as compared to As(III) which is highly mobile and toxic (Bhattacharyya & Gupta 2008). It has been found that As(III) removal is optimum at a pH range of 4–6 while As(V) removal is optimum at a pH range 4–8 (Mudzielwana et al. 2020).

In aqueous systems, chromium usually exists in both trivalent Cr(III) and hexavalent Cr(VI) oxidation states. Cr(III) is about 300 times less toxic, less mobile and less bioavailable than Cr(VI) (Krishnani & Ayyappan 2006). The main sources of Cr(VI) are leather tanning, the mining of chrome ore, the production of steel and alloys, etc. At an acidic pH (under 3.6), Cr(III) is the prevalent species. The most common conventional method for Cr(VI) removal is the reduction to Cr(III) at pH 2.0 and precipitation of $Cr(OH)_3$ by increasing the pH to 9–10 using lime. The disadvantage of precipitation is the disposal of the solid waste (Miretzky & Cirelli 2010). (Krishnani & Ayyappan 2006) have reported that many agriculture by-products (sawdust, paper mill discard, sugarcane bagasse, wheat bran, wheat straw, corn stoves, etc.) to be good low-cost adsorbents for the removal of both trivalent and hexavalent chromium from water. The efficient removal of both Cr(III) and Cr(VI) was confirmed by sorption on the *Ectodermis* of *Opuntia* (Barrera et al. 2006).

Oxidation system with cascade aeration and subsequent leaching on the bed. A cascade system is used to oxidise anoxic mine effluent. Cascade oxidation is commonly used in passive systems. However, its effectiveness is commonly assumed and rarely measured. Cascade oxidation of mine water with a pre-diffusion aerator was experimentally tested in the field. This combination has been shown to be suitable for the oxidation of Fe(II) (Oh et al. 2015).

Cascade oxidation can be supplemented by bed leaching to remove metals. CaO containing slag is preferred for Mn removal (Trumm & Pope 2015). The slag is used in the treatment of a mine effluent with a pH > 8 (Ziemkiewicz & Skousen 1999; Simmons et al. 2002; Skousen 2002).

Wetlands. Wetlands are passive systems. This system is used to remediate contaminated water by the synergy of plants, substrates and microbes (Jia et al. 2020; Zhang et al. 2020). In wetlands, NMDs are treated by the adsorption and precipitation of heavy metals and metalloids on substrates in the wetlands and assimilation by plants (Wang et al. 2021). Therefore, in the design and operation of the constructed wetlands, the appropriate choice of substrate materials in accordance with the properties of the inflow wastewater is crucial. Wetlands have a positive effect on the quality of mine water due to the removal of contaminants by adsorption, coprecipitation and colloid filtration (Sracek et al. 2018). The effectiveness of remediation in wetlands has been confirmed by several studies (Etteieb et al. 2021; Nguyen et al. 2021; Chen et al. 2022).

Sorption methods. Metal sorption is a rapid process that can be used in passive treatment systems to reduce the concentration of metals/metalloids in contaminated neutral drainage below regulatory thresholds (Calugaru et al. 2018). Adsorption and binding of the adsorbate to the adsorbent is preferred at a specific pH. At an acidic pH, a great number of H^+ ions in the solution compete with metal ions for active sites on the adsorbent and, consequently, the metal removal is less. With an increasing pH, H^+ ions are reduced (Esmaeili et al. 2019). Thus, a change in the pH may reverse the reaction (Vakili et al. 2019).

The main advantages of adsorption are the operational simplicity, a variety of the adsorbent materials, no need for any other chemical substances and little sludge production (Božić et al. 2013; Esmaeili et al. 2019).

The key issue is the financial complexity of the sorption process. High financial costs arise during the actual transport of the sorbent to remote locations. Previous research has confirmed good sorption capacities in the sorption of metals on various low-cost, natural materials and industrial waste products, such as natural clay (Esmaeili et al. 2019), recycled concrete aggregates, fly ashes (Jones & Cetin 2017), compost (Richard et al. 2020), raw and modified wood ash (Calugaru et al. 2020), raw and modified (by thermal activation) dolomite (Calugaru et al. 2016), a natural zeolite (Wingenfelder et al. 2005), iron filler (Sekula et al. 2018), and dolomite (Ayoub & Mehawej 2007; Kocaoba 2007; Pehlivan et al. 2009; Salameh et al. 2010).

With the operating parameters, such as the pH, ionic strength, and sorbent, the sorbate ratios vary widely between studies, making comparisons difficult (Richard et al. 2020a).

The choice of the adsorbent itself also depends on the element that one wants to remove during the remediation of the mine effluents. With this in mind, iron filler (Sekula et al. 2018), hydrothermally activated wood ash (Calugaru et al. 2020), iron-containing biochar (Calugaru et al. 2019), dolomite (Ayoub & Mehawej 2007; Kocaoba 2007; Pehlivan et al. 2009; Salameh et al. 2010), modified, e.g., thermally activated dolomite (Walker et al. 2005), and a natural zeolite (Wingenfelder et al. 2005) have been used as adsorbents in the past.

It is also necessary to consider the sorption capacity of the sorbent for a particular metal. The high sorption capacity of metals has been confirmed for Mn oxides prevalent in soils and natural waters (Sánchez-España & Yusta 2019).

An important factor in selecting a suitable sorbent for metal/metalloids sorption is to consider the properties of the NMDs in a particular area. Indeed, previous research on a particular sorbent may focus on one major area (e.g., the United States) (Fuller & Harvey 2000; Kay et al. 2001, Lee et al. 2002; Tani et al. 2004) and, therefore, the results of the sorption may not be relevant in the conditions of other localities (e.g., Europe).

Adsorption systems can be implemented with and without a pH adjustment. These disposal methods offer several advantages in terms of the design, operation and efficiency compared to other wastewater treatment technologies (Rodríguez et al. 2020). The removal of hazardous elements by sorption depends on the pH of the solution. Metal cations, such as Pb, Cu, Zn and Cd, are more easily sorbed at neutral to moderately alkaline pH values (Dzombak et al. 1991; Gibert et al. 2005). Biosorption technologies are currently being investigated. The efficiency of the process yields quite satisfactory results (Aryal et al. 2010; Pinto et al. 2011; Westholm et al. 2014; Richard et al. 2020b)

Passive reactor system. Passive reactors (PRs) are considered to be an efficient technology used to neutralise acidity and remove metals/metalloids and sulfates from MDs. The principle of this technology lies in the gravitational flow of the MD through the reactor, where contact with the reactive mixture promotes treatment by various physical, chemical and biological mechanisms (Dvorak et al. 1992). The removal of metals consists in their precipitation as the pH increases. This effect can be achieved by reacting a closed system with calcite in a single gravity, sealed bed of limestone gravel (anoxic limestone runoff (ALD)) (Nuttall & Younger 2000).

From an economic point of view, it is necessary to focus on cheap materials when choosing a reactive material for the NMD treatment. Compost, ash, waste shell material, iron ochre and a mixture of blast furnace slag (BFS) and basic oxygen furnace slag (BOS) can be used as inexpensive reactive materials (Warrender et al. 2011).

For the successful removal of metals (e.g., Cu, Pb, Zn, Hg and Cd), waste materials can be used as a cheap alternative to more expensive reactive materials. Useful reactive waste materials include wood, chitin, coconut shells, clay soil, and seaweed (Bailey et al. 1999; Pearce et al. 2007).

Precipitation. Precipitation is a conventional method developed for removing and recovering metals/metalloids from solutions. Precipitation from aqueous solutions contaminated with metals/metalloids can be achieved by the formation of their respective (*i*) sulfide, (*ii*) hydroxide and sometimes (*iii*) carbonate salts (Sethurajan et al. 2017). Several elements, such as arsenic, can also precipitate during flocculation with iron and aluminium salts. The efficiency of the regeneration by precipitation very much depends on the initial concentrations in the solution and on the pH of the system. The main disadvantages of precipitation processes are the high requirements for chemicals to adjust the pH and the formation of poorly settled and dewaterable sludge containing toxic compounds (Ahalya & Ramachandra 2003).

Precipitation occurs when the concentration of ions in the solution exceeds the solubility product (Wang & Starnik 2005), and can be caused by changes in the ionic equilibrium of the system with the addition of reaction products (either metals, sulfides or hydroxides). Metal precipitation consists of different stages: (*i*) nucleation, (*ii*) nuclear growth, and (*iii*) aggregation or crystallisation (Benning & Waychunas 2008).

Hydroxides (NaOH and $Ca(OH)_2$), carbonates or some inorganic sulfur compounds can be used for precipitation – Na_2S, NaHS, $(NH_4)_2S$ and H_2S, which are then used to precipitate metal sulfides (Sethurajan et al. 2017). Sulfide precipitation is advantageous because metal sulfide precipitates are less soluble, the reaction rates are faster, the settling properties are better, and sulfide precipitates could be combined with ores in metallurgical processes (Lewis 2010). Non-selective precipitation methods are effective for most metals, but it is difficult to meet the requirements of discharge standards with this method only. In addition, the post-precipitation treatment has many practical limitations, such as causing secondary contamination due to the addition of large volumes of alkalis and high reagent costs (Zhu et al. 2019).

Biological oxidative precipitation. This technology can be used in passive treatment systems. In addition to neutralising contaminated water (in the case of acid mine effluents), passive limestone beds also allow the biological oxidation of metals (such as Mn (II)) (Johnson & Younger 2005; Means & Rose 2005). A wide variety of bacteria, fungi and algae can be used for the enzymatic oxidation of metals/metalloids (Tebo et al. 2004; Thompson et al. 2005).

At a neutral pH, Mn (II) can be oxidised by the microbiological activity or surface-catalysed heterogeneous oxidation on Mn (III / IV) oxide surfaces (Junta & Hochella 1994; Tan et al. 2010).

Membrane separation. Reverse osmosis (RO), a high-pressure membrane separation process, can be used to regenerate metals/metalloids over a wide pH range of 3–10 (Kang et al. 2000). Ligand hollow fibre membranes have shown the potential to regenerate Sb over a wide pH range. The hollow fibre chelating membrane contains an iminodiethanol (IDE) group as a chelator (Nishiyama et al. 2003). Recently, RO has been used to treat industrial wastewater from field mining operations in Victoria-Australia with an average extraction efficiency of 10% for Fe^{3+}, 48% for Zn^{2+}, 82% for Ni^{2+}, 66% for As^{3+} and 95% for Sb^{3+} (Samaei et al. 2020).

Ultrafiltration is used at low transmembrane operating pressure. Because the pores of the ultrafiltration membrane may be larger than heavy metal ions, additives may bind to the metal ions to increase the size of the metal ions. Therefore, micellar enhanced ultrafiltration has been proposed, which is created by combining ultrafiltration and a surfactant. Micellar enhanced ultrafiltration has high flux and high selectivity, leading to low energy consumption, high removal efficiency and less space, and is, therefore, most suitable for waters with low heavy metal concentrations (Huang et al. 2017; Rahmati et al. 2017).

Nanofiltration membranes are composed of polymer composites of multiple-layer thin-film of negatively charged chemical groups (El-Sherif et al. 2013). Anti-fouling nanofiltration membranes were synthesised through phase inversion and used to extract Fe^{3+}, Al^{3+}, Co^{2+}, Cd^{2+}, Cu^{2+}, and humic acid from wastewater and reached an extraction efficiency between 94 and 98% (Jamil et al. 2018).

Microfiltration (MF) employs a microporous membrane to remove micron-sized particles, bacteria, viruses, protozoa, contaminants, pollutants. The application of MF in heavy metal removal has not drawn enough attention because of its low removal ability. However, it has been used by modifying the membrane or chemical pre-treatment of the feed solution (Qasem et al. 2021).

Electrodialysis is used to separate ions at the expense of the electric potential difference. Electrodialysis uses a series of cation exchange membranes and anion exchange membranes, alternatively arranged in parallel, to separate ionic solutes (Abdullah et al. 2019).

Electrodialysis has been used to separate Ni^{2+}, $Pb^{2+,}$ and K^+ from synthetic solutions to attain an extraction efficiency of 96.9%, 99.9%, and 99.9% for Ni^{2+}, $Pb^{2+,}$ and K^+, respectively (Nemati et al. 2017). A pilot-scale electrodialysis system has also been used to extract Cu^{2+}, Ni^{2+}, and traces of Cd^{2+}, Fe^{3+}, Cr^{6+}, and Zn^{2+}, and exceeded a 90% removal rate (El-Sherif et al. 2013). The main advantages of electrodialysis include a high separation efficiency of positively and negatively charged ions, coping with low metal/metalloids concentrations, and a low operating pressure. The main disadvantages of electrodialysis include its high energy consumption, separation efficiency influenced by operational parameters and low selectivity (Arana Juve et al. 2022).

Membrane distillation and liquid membranes have also been used for wastewater treatment. A membrane distillation process has been reported to achieve over a 96% removal of Ca^{2+}, Mg^{2+}, Fe^{3+}, and Fe^{2+} (El-Sherif et al. 2013) and more than 99% for As^{3+} and As^{5+} (Qu et al. 2009).

Combination of aerobic and anaerobic treatments. This combination is intended for the treatment of an almost neutral to slightly alkaline mine water. In the aerobic treatment step, the removal of metals (e.g., Mn) takes place. Wetland systems can serve to accomplish this step (Batty et al. 2008; Ye et al. 2001). The subsequent anaerobic stage serves for the bacterial reduction of SO_4^{2-}. This system is recommended for implementation in a flow-through design containing an organic substrate only after the removal of Fe and other metals (Hedin et al. 1994).

Methods of metal/metalloids recovery from neutral mine effluents

Metal recovery is mainly used to ensure the economic efficiency of the whole strategy, especially when it comes to buying expensive inputs (adsorbents, chemicals).

Leaching with acidic solutions. Acidic solutions can be used to recover metals/metalloids after the sorption process. The desorption mechanism is similar to the ion exchange, where metals are eluted from the sorbent with a suitable solution to obtain a small, concentrated metal-containing solution. The regeneration of metals from the sorbent can be achieved by relatively inexpensive acids – HCl, HNO_3 and H_2SO_4 (Tsezos 1984; Kuyucak & Volesky 1989; Aldor et al. 1995) (Table 4).

Hydrochloric acid can desorb Cu, Zn, Cd, Co with an efficiency of 85–90% (Jiao et al. 2017; Kołodyńska et al. 2017). Desorption of Fe, Al, Zn, Ni and Pb metals is more efficient in an acidic environment of 76–98% during the first 3 hours compared to a neutral environment of less than 5% (Sahoo et al. 2013). Experimental experiments with HCl have shown that the too low pH required for efficient recovery causes the sorbent to dissolve. Therefore, it is no longer possible to regenerate this sorbent in the next step of the process (Aryal et al. 2010). Nitric acid is a better desorbent (compared to HCl and H_2SO_4) for the desorption of Cu, Zn, Co, Cd, Pb and can be used for up to 4 desorption cycles in a row (Chatterjee & Abraham 2019). Sulfuric acid (inorganic) is able to desorb 32.1% Cu and 20.3% Cd (Kamaruzaman et al. 2017), but is not suitable for lead desorption (Chatterjee & Abraham 2019).

In addition to the acids already mentioned, metal ion leaching has been performed in the past using ethylenediaminetetraacetic acid (EDTA) to recover metals (Sun et al. 2001; Xueyi & Inoue 2003). In contrast to HCl, it is possible to recover metals with an EDTA solution with a successful biosorbent regeneration process - macroalgae and microalgae (Zhou et al. 1998; Tan et al. 2002; Amorim et al. 2003).

Leaching with ultrapure water. Ultrapure water leaching can be used to desorb metals after sorption. However, ultrapure water does not have a high sorption efficiency of metals compared to other leaching agents (e.g., acidic and basic solutions) (Li et al. 2009).

Leaching with alkaline solutions. Diluted solutions of basic salts, such as NaOH, $NaHCO_3$, are mostly used as desorbents (Chatterjee & Abraham 2019). Among the alkaline solutions, NaOH and NH_4OH solutions are the most effective and often used for the desorption of ions from adsorbents. The desorption mechanism by basic processing involves deprotonation and negatively charged adsorption sites in the adsorbents. These characteristics weaken the existing electrostatic interactions between the adsorbent functional groups and the metal/metalloid ions and the subsequent separation of the adsorbed ions from the adsorption sites (Vakili et al. 2019). NaOH can be used to desorb As, Se, Cr, Ni and Co (Kamala et al. 2005; Zhang et al. 2009; Ipeaiyeda & Tesi 2014). Alkaline solution stripping has proven to be a suitable technology for regenerating the biosorbent with the adsorbed element (As) (Boddu et al. 2008).

Alkalis are effective desorbents for desorbing metals/metalloids from chemical adsorbents or chemically modified adsorbents, acids are effective for desorbing bioadsorbents and the chelating agent EDTA is the most effective desorbing agent for biomass desorption (Lata et al. 2015).

Salt desorption. Salts can be considered as suitable desorbents for the regeneration of adsorbed metal/metalloid ions because they have the ability to weaken the interaction between the ions and the binding sites on the surface of the adsorbent to form stable complexes. Salts can be used to desorb heavy metals: NaCl, KNO_3, Na_2CO_3 and Na_2SO_4 (Vakili et al. 2019). The desorption mechanism may be the deprotonation of adsorbent functional groups, which releases heavy metal ions. In addition, the strong power supply of these desorbents readily desorbs adsorbed metal ions and forms a stable complex with the desorbents (Javanbakht et al. 2016; Vakili et al. 2019).

Selective precipitation. In this process, the metals/metalloids are obtained by several precipitation

Table 4. Critical evaluation of the metal recovery methods

Recovery methods	Advantages	Disadvantages	References
Leaching with acidic solutions	faster and more efficient (compared to alkaline leaching and water leaching) acidic conditions support the dissolution of Fe and Al and, thus, the release of adsorbed/precipitated metals most suitable for the desorption of bioadsorbents	consumption of leaching acids along with their price low pH may cause dissolution of adsorbent or is not suitable for repeated use	Aryal et al. (2010); Chatterjee andAbraham (2019); Li et al. (2009); Calugaru et al. (2020)
Leaching with alkaline solutions	most suitable for desorption of chemical or chemically modified adsorbents can effectively desorb heavy metal anionic compounds such as $H_2AsO_3^-$, CrO_4^{2-}, $HCrO_4^-$, $HCr_2O_7^-$ and $Cr_2O_7^{2-}$	some metals are better desorbed at slightly acidic pH consumption of leaching alkaline solutions along with their price	Boddu et al. (2008); Chatterjee and Abraham (2019); Vakili et al. (2019)
Salts desorption	formation of stable complexes electrostatic forces allow desorption of metal ions allows better adsorbent regeneration	consumption of leaching salts along with their price lower desorption efficiency compared to acidic and alkaline leaching	Javanbakht et al. (2016); Chatterjee and Abraham (2019); Vakili et al. (2019)
Leaching with ultrapure water	does not burden the environment	low efficiency	Li et al. (2009); Aryal et al. (2010)
Selective precipitation	reduction of the volume of sludge formed and possible recovery of metals metals can be selectively obtained	for good efficiency is necessary to ensure mixing consumption of precipitation chemicals	Wei et al. (2005); Yan et al. (2015)
Sludge electrolysis	a wide amount of metals can be recovered large number of use cycles no additional chemicals need to be supplied	necessary removal of residual sludge energy and economic intensity	Yi et al. (2019); Jin and Zhang (2020)

stages. The precipitate is concentrated and filtered under reduced pressure. The filtrate is pumped into a seed tank and the dehydrated precipitate is stored for further purification. This technology has been proven to be effective in recovering metals from mine effluents. The advantage of the process is that the sludge can be reused after precipitation, thus creating no residue (Yan et al. 2015). In this technology, it is important to consider the pH values. A pH range of 6.0–7.0 is recommended for Al recovery. During Fe recovery, the pH must be adjusted to acidic values (3.5–4.0) (Wei et al. 2005).

Bioelectrochemical systems. The basics of bioelectrochemical systems (BESs) are based on the metabolism of exoelectrogenic microorganisms capable of catalysing electrochemical reactions taking place on the electrode surfaces of an electrochemical cell (Ren 2013). Metal ion recovery separately from waste streams via BES has the advantage of lower energy consumption compared to established clean electrochemical technologies (Wang & Ren 2014). BESs are highly efficient, consume less energy, and are safe for the environment (Yang et al. 2019).

Electrolysis. Promising results obtained with the Chemelec Cell and HSA reactor systems in recovering metals from dilute process streams suggest that this technology may also be applicable to the recovery of metals such as Cu, Cd and Zn from mine effluents (Dinardo et al. 1991) (Table 5). Electrolysis has also been used to successfully recover Mn and Li from waste streams (Jin & Zhang 2020). Yi et al. have shown that in addition to a large number of metals for recovery (Al, Ni, Cu, Pb, Ag, Pd, Pt and Au), the sludge electrolysis method is able to reuse the sludge in up to 13 cycles. Sludge electrolysis requires additional sludge removal with acid (HCl) (Yi et al. 2019).

Table 5. Critical evaluation of combination metal removal and recovery

Treatment method	Recovery method	Advantages	Disadvantages	References
Adsorption on dolomite	leaching	cheap and available adsorbent suitable for sorption of Zn, Fe and Mn capacity can be increased by calcination or thermal activation can be used in long cycle with long retention time	can be used in only one cycle problematic regeneration (salts are best used for desorption, adsorbent dissolves when using acid)	Ivanets et al. (2016); Calugaru et al. (2020, 2021)
Adsorption on fly ash	leaching with acidic solutions	cheap and ecological adsorbent suitable for sorption of Zn, Fe and Mn by leaching in acidic solutions, the adsorbent can be regenerated very well	fast depletion of adsorbent (more frequent adsorbent change required)	Sahoo et al. (2013); Saha and Sinha (2018)
Membrane separation	electrolysis	high selectivity and resource recovery pure metals can be obtained by electrolysis it is possible to obtain water and heavy metal concentrate	a great deal of pre-treatment is required pore-blocking weak antibacterial properties requires maintenance expensive	Yi et al. (2019); Samaei et al. (2020); Jin and Zhang (2020)
Precipitation by LDH	electrolysis	high efficiency of Zn, Mn, Pb, Ni and Cd removal by electrolysis it is possible to obtain water and heavy metal concentrate	worse sludge handling (it is more diluted than the one arising in passive systems) expensive chemical consumption	Frau et al. (2020); Yi et al. (2019); Jin and Zhang (2020)

LDH – layered double hydroxides

Proposal of strategies for the treatment of neutral mine drainages from selected localities in Slovakia

In the Štiavnica-Hodruša ore district, the main sources of mine water are the adit Zlatý stôl, the Nová odvodňovacia štôlňa adit, the Voznická dedičná štôlňa adit and the drainage from the sludge reservoir in the village of Hodruša-Hámre. All the discharges contaminate the surface waters and despite the neutral pH, can be a source of potentially hazardous elements. Of the determined elements, the highest concentrations of Zn, Mn and Fe are reached, the removal of which must be focused on when designing the strategy.

The use of limestone/dolomite is a widely used method of MD remediation. When used in AMD, limestone dissolves, which increases the pH and alkalinity. Increasing the alkalinity causes a reduction in the concentration of metals through oxidation, hydrolysis and precipitation. Limestone/dolomite drains can also be used for the partial sulfate treatment in the MD, either by gypsum precipitation or by sorption of sulfates on the limestone surface (at lower sulfate concentrations) (Calugaru et al. 2021).

Ash is a cheap, ecological and unconventional alternative to activated carbon. It can remove various elements such as Fe, Al, Mn, Ca, Mg, Zn, Cu, Cd, Ni, Co. It can be used as an alternative to dolomite and limestone. Treatment of mine water with fly ash also reduces the Mn concentration below the detection limit (Saha & Sinha 2018).

Both anionic and cationic contaminants can be removed by precipitation with layered double hydroxides (LDHs). LDH precipitation generally assumes neutral alkaline conditions and the presence of dissolved divalent and trivalent metals in appropriate proportions. The method is very simple and is based on two steps, which involve the addition of an Al salt (lowering the pH) and using a base (e.g., NaOH) to restore the pH of the solution to a neutral or slightly basic value. The metal removal efficiency is generally very high (up to 100% Zn, 98% Mn, 96% Pb, 99% Ni, 40% Cd) (Frau et al. 2020).

The simplest way is probably to use adsorption, which is not too expensive and, at the same time, a wide range of readily available adsorbents can be used. It also does not require regular maintenance and many adsorbents can be used over a long cycle.

This method was applied *in situ* in the locality of Poproč, Slovakia for the treatment of NMD flowing out of the adit Agnes. The mine water flowed through two reactors with an adsorbent each with a volume of 1 m^3, the retention time in each reactor being approximately 2.2 h. The system was in operation for 2.3 years from May 2013 to August 2015. After approximately 1.25 years, one reactor had to be cleaned and put back into operation due to overflowing with ochre precipitates. The average efficiency of the Sb and As removal from the mine water was 84% and 89% over a period of 2.3 years, respectively (Sekula et al. 2018).

In the case of remediation of NMDs in the Štiavnica-Hodruša region, it would be necessary to find the most suitable adsorbent by laboratory experiments and subsequently verify its effectiveness *in situ*. However, even with such a simple method, it will be necessary to perform a minimal inspection of the equipment directly in the field. In the case of metal recovery, it will be necessary to determine the amount of trapped metals in the adsorbent and verify the profitability of their recovery.

CONCLUSION

Neutral mine drainages have a significant presence in Slovakia. Not only the pH levels, but also the concentrations of potentially hazardous elements must be taken into account when treating NMDs. The mining site itself has a significant impact on the mine effluent properties. From the point of view of simplicity of implementation, passive methods still have a dominant position over active methods of NMD treatment. The main advantage of passive methods lies in the easier implementation and lower costs compared to active technologies. Active methods, on the other hand, offer higher metal removal efficiencies. The choice of a suitable treatment method should not only take the efficiency of removing contamination into account (e.g., heavy metals), but also the economic costs of the process, with respect to the critical evaluation of the methods from a comprehensive point of view. In context of the economic efficiency, it is important to focus on a suitable combination of the treatment method (specific for a particular mine water) with the methods of recovery of heavy metals. Metal removal and the recovery processes can be complemented positively (stripping using NaOH without sorbent dissolution in the metal recovery process after sorption) or negatively (sorbent dissolution in leaching in an HCl solution for metal recovery). BES technologies are offered as new methods of metal recovery, the attractiveness of which lies primarily in the energy savings. The necessary criterion is the feasibility of a remediation strategy directly in the locality. In the future, research in the field needs to focus on the analysis of the negative aspects of the environment that may disrupt, with respect to support the implementation of the selected method of remediation.

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