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MINING AND THE ENVIRONMENT - UNDERSTANDING PROCESSES, ASSESSING IMPACTS AND DEVELOPING REMEDIATION

The use of low-cost adsorbents for wastewater purification in mining industries

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Abstract Recently, great attention has been paid to the environmental problems in mining industry. At present there are different ways of mineral processing, as well as various methods of wastewater treatment, most of them are expensive. Work is ongoing to find low-cost treatments. In this article, low-cost adsorbents, potentially useful for wastewater treatment on mining and metallurgical plants, are reviewed; their characteristics, advantages, and disadvantages of their application are compared. Also adsorption of different metals and radioactive compounds from acidic environment similar to composition of mining and metallurgical wastewaters is considered.

Keywords Wastewater treatment · AMD · Low-cost adsorbent · Mining industry

Notation

AMD	Acid mine drainage
DMC	Dairy manure compost
EDTA	Ethylenediaminetetraacetic acid
DTPA	Diethylenetriaminepentaacetic acid
q_e	Equilibrium adsorption capacity
q_m	Maximum adsorption capacity
K	Coefficient of Henry's law
C	Concentration
C_r	Reduced concentration ($C_r = \frac{c}{c_{sat}}$), where C_{sat} is saturation concentration
A, B, D	Empirical parameters
k_0	Shaper parameter
b_0	Affinity coefficient
n	The parameter characterizing the type of pores
M_1, M_2	Subscript denoting the different pore size distribution

Introduction

Since the beginning of the technical revolution in the mid-nineteenth century, the rapid development process and improvement of life quality were accompanied by environmental degradation and as a result the deterioration of human health, the emergence of new diseases associated with poisonous waste production, as well as different co-evolutionary processes in aquatic system (Paavola 2011). At present, the technical process continues to evolve forcing one to think about tightening of measures to protect the environment. In particular, the extraction of minerals leads to serious disturbances in the environment of the surrounding areas during and after operation.

One of the modern trends in mining and metallurgical industries is the reduction of waste production. Whereas a mining industry without waste generation is impossible yet, it is necessary to find ways to reduce and minimize generation of waste (Chan et al. 2008; Hilson 2003; Hilson 2000; Silva et al. 2010; Wolkersdorfer 2005; Wolkersdorfer 2008; Zhong-ning et al. 2009). "Between 2000 and 2002, the Mining, Minerals and Sustainable Development (MMSD) project carried out research, analysis and consultation" (Buxton 2002). According to summary data presented in the article (Azapagic 2004), less than half of mining

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companies followed MMSD plan. Economic and social aspects of improvement of mining technology are presented. Also the demand to improve current technologies to reduce waste production, maximize efficiency of natural resources extraction, and reuse of the necessary resources for these areas (in particular water recycling) are mentioned (Azapagic 2004; Driussi and Jansz 2006a; Driussi and Jansz 2006b; Huang et al. 2010; Suppen et al. 2006). Kolmert and Johnson (2001) discussed the possibility of water recycling in mining industries using pre-biological treatment. Many mining industrial countries try to implement low and as a future plan even zero (Australia) waste mining policy (Amankwah and Anim-Sackey 2003; Azapagic 2004; Driussi and Jansz 2006a; Driussi and Jansz 2006b; Garcia et al. 2001; Hilson and Murck 2001; Ntengwe 2005; Sarrasin 2006; Suppen et al. 2006). Mining companies show the desire to cooperate with universities and research institutions in order to improve their current technologies and the environment situation (Amankwah and Anim-Sackey 2003; Garcia et al. 2001; Hilson and Murck 2001; Sarrasin 2006; Solomon et al. 2008; Suppen et al. 2006). Innovative design is one of the approaches that increase the production capacity up to 30–90 %. Thus a new method of solvent-extraction electrowinning (SX-EW) applied in 80–90 years of twentieth century made possible to use low-grade and oxidized ores that were previously considered as wastes (Garcia et al. 2001).

Impact of mine wastes to human health

According to the Protection of the Marine Environment of the North-East Atlantic (the OSPAR Convention), the total global emission of toxic metals in the atmosphere made up from 1990 to 2008 amounted to 107 tonnes of cadmium, 37 tonnes of mercury, and 1,109 tonnes of lead (OSPAR 2008).

These are just some elements that enter the atmosphere as a result of human activities. The chemical pollutants enter human body in different ways: directly with inhaled air and water and indirectly through food chains from soil, water, and air, as some plants and animals are able to bioaccumulate toxicants. This leads to various human body malfunctions and diseases (Bermudez et al. 2011; Silva et al. 2010). Ions Pb^{2+}, Hg^{2+}, CH^3Hg^+, Co^{2+}, and Cd^{2+} form stable complexes with amino acids and other biomolecules containing thio-(HS-) or alkiltiogroups (RS-). Many metal complexes with organic ligands are similar in their structure (geometry, charge distribution in the molecule, etc.) to amino acids, hormones, and neurotransmitters in human body and can, therefore, communicate with the relevant receptors, so called effect of mimicry. Another important mechanism of toxic action of metals entering the human body from waste production is to replace metals (essential for organic synthesis) to toxic metals in their complexes. This leads to loss of biological activity of cells containing corresponding complexes (Isidorov 1997). For example, aluminium replaces calcium in human bones, which causes bone disease (Dhaese et al. 1996). By acting on nervous system, it is one of the causes of Alzheimer's disease (Kekovic 2010).

For example, "itai-itai" disease ("ouch-ouch" in English), took place in Japan in the 40–60 years of twentieth century. The issue arose with the residents who had been consuming rice containing cadmium (1 gkg^-) for a long time. This rice was grown in the fields irrigated with cadmium-contaminated water, the irrigating waters were mixed with wastewater from cadmium mining and ore dressing refinery process (Makoto 1991; Yoshida 1999). Consumption of fish containing methylmercury in high concentrations causes the "Minamata disease" (Cortes-Maramba et al. 2006; Ekino et al. 2007). Arsenic is a human carcinogen (Hamilton 2000; Xu 2012) and also may cause hypertension (Abhyankar 2012). Toxic effects of other elements on human health are also studied (Broadway et al. 2010; Newhook et al. 2003; Wang et al. 2011). For the past 100 years cyanides are mainly produced for gold and silver mining industry. The reactions, known as Elsner's Eqs. 1 and 2 (Senanayake 2006), are describing silver and gold cyanidation processes:

$$4Ag + 8CN^- + O_2 + 2H_2O = 4Ag(CN)_2^- + 4OH^- \tag{1}$$

$$4Au + 8CN^- + O_2 + 2H_2O = 4Au(CN)_2^- + 4OH^- \tag{2}$$

Cyanides are extremely toxic for humans. Cyanide poisoning is a form of histotoxic hypoxia because organism cells are unable to use oxygen, primarily through inhibition of cytochrome c-oxidase. Inhalation of high concentration of cyanides causes coma with seizures, apnea, and cardiac arrest, with death following in a matter of minutes. At lower doses, loss of consciousness may be preceded by general weakness, giddiness, headaches, vertigo, confusion, and perceived difficulty in breathing. At the first stages of unconsciousness, breathing is often sufficient or even rapid, although the state of the sufferer progresses towards a deep coma, sometimes accompanied by pulmonary edema, and finally cardiac arrest. Skin color goes pink from cyanide–hemoglobin complexes. A fatal dose for humans can be as low as 1.5 mg/kg body weight (Deutsch 2012).

Sources of mine wastes

There is ground and underground mining. According to European Commission, the ore extraction capacities are generally much lower for underground mining method than for surface quarries. However, the quantity of waste produced per unit of ore mined is much lower, and the ground area of this type of underground mine is considerably smaller than for surface quarries (Co-ordination by Charbonier B 2001). At present surface mine is approximately 80 % of all world mining.

Mining and ore processing generate solid and liquid wastes. This work is considering a liquid mine wastes (wastewaters and solid wastes from different industries) as potential materials for wastewater purification. The sources of contaminated minewater from mining operations are drainage from underground workings, runoff from open pit workings, waste rock dumps from mining activities, mill tailings, ore stockpiles, spent ore piles from heap leach operations (Brown et al. 2002).

General diagram showing possible ways of wastewater appearance with ore-processing industries, surface and underground mine is shown on Figure 1. It can be seen from this scheme, that pollution of surface and groundwaters, as well as air takes place during the active mining. Composition of wastewaters from mine, metallurgical, and ore enriching plants can vary greatly depending on composition of the solid as well as methods of extraction and enrichment. Mine wastewaters can be categorized into three groups according to their acid–base properties: acid mine drainage (AMD) with pH at 6 and below, neutral mine drainage with pH 6 and above, and saline mine drainage with pH above 6 containing more than 1,000 $\mathrm{mg\,L^{-1}}$ carbonates (Wolkersdorfer 2008).

Natural oxidation of sulfide minerals, together with reactions of the base minerals in the rock, which are exposed to air and water, is forming acid rock drainage. Activities involving excavation of rock with sulfide minerals, such as mining, accelerate the process due to increase in exposure of sulfide minerals to air, water, and microorganisms. In this case the process name is acid mine drainage. The drainage produced from the oxidation process may be neutral to acidic, with or without dissolved heavy metals, but such drainage always contains sulfate (Wolkersdorfer 2008).

The high acidity of groundwater and their toxic metals contaminations are confirmed by many researchers (Ahmaruzzaman 2011a; Akcil and Koldas 2006; Barakat 2011; Evangelou 1998; Fang et al. 2007; Huang et al. 2010; Jung et al. 2012; LEI et al. 2010; Pahlavanzadeh et al. 2010; Rodrigues Silva 2009; Sivulka et al. 2007; Tuazon et al. 2012). It happens due to leaching of metals from the tailings. As it was mentioned in Akcil and Koldas (2006): AMD is produced when sulfide-bearing material is exposed to oxygen and water; however, one should remember that not only iron sulfides produce AMD but many other sulfides as well. The acidity of waters varies depending on the season, consequently the changing of concentration of metals (Cu, Pb, Cd, As, Zn, Al, Fe, Mn) is observed (the lower the acidity, the higher the concentration) (Lei et al. 2010). Bacterial activity increases the rate of this process (Akcil and Koldas 2006). The AMD is the main problem of all mining industries. Hilson and Murck (2001) describe the potentially serious environmental problems in the gold-mining industry such as sedimentation, erosion, and habitat destruction, and discuss ways to prevent them. Main pollutants in the extraction of gold-mining industry are cyanide contamination and AMD, as uranium, because it is a "companion" in the deposits of gold and silver. While in some countries, such as the US and Canada, mercury amalgamation was replaced with cyanide technologies, in many other countries the old method is still in use, the mercury contamination (Amankwah and Anim-Sackey 2003; Hilson and Murck 2001).

Treatment methods of mining wastewaters

The composition of AMD can vary depending on the rock, as each mine is unique in its composition (Akcil and Koldas 2006). Therefore the individual approach to AMD treatment is necessary, but there are general approaches to it.

All methods of wastewater treatment can be divided into two main groups: active and passive. This classification is described in details in Brown et al. (2002) and Johnson and Hallberg (2005).

Active wastewater treatment consists of mechanical, chemical, mechanochemical, biochemical, and physicochemical methods. Detailed discussion about the stages of wastewaters treatment can be found in comprehensive review by Gupta (2012). Below step by step diagram of wastewater purification is shown.

Mechanical treatment (Kavindra 2011) is used for removal of solid substances and contaminants in the colloidal state. Since in mining and metallurgical wastewaters concentration of suspended solids is usually high (Verma et al.), mechanical treatments are widely used. Chemical treatment involves chemical reactions that help to remove impurities from solutions. For example neutralization of AMD before metals are removed (Akcil and Koldas 2006). Redox reactions (Søndergaard 2009), precipitations (Fausto 1986), and reactions accompanied by gas formation are used as well. Biochemical wastewater treatment is oxidation of organic contaminants by microorganisms capable to carry out mineralization of organic compounds (Mandal et al. 2010). Physicochemical methods of wastewater purification include sorption, extraction (Kuan et al. 2010; Li et al. 2004; Álvarez et al. 2002), coagulation (Gao et al. 2007; Ren et al. 2011), electrolysis (Curteanu et al. 2011; Jenke and Diebold 1984), catalysis (Shon et al. 2008), ion exchange (Bochenek et al. 2011), magnetic separation (Ambashta and Sillanpää 2010), crystallization (Bian et al. 2011) etc. Usually physicochemical methods are applied in combination (Agustina

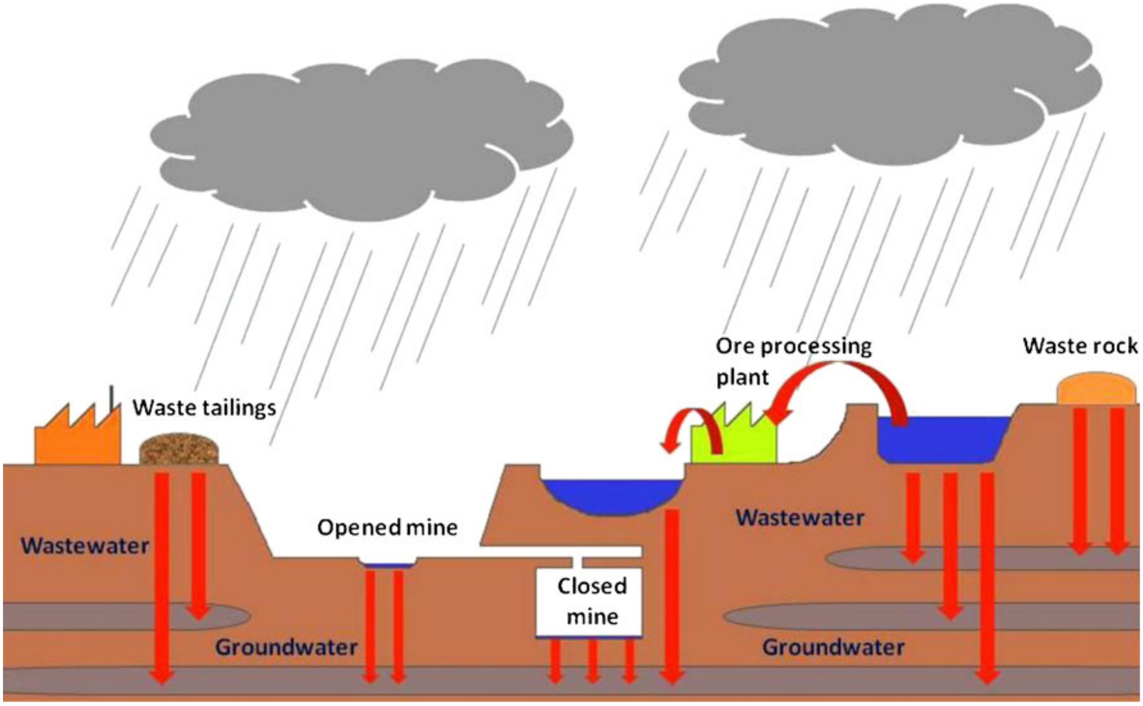

Fig. 1 Pollutant transfer

et al. 2005; Vimonses et al. 2010; Zhang et al. 2006; Zhang et al. 2007).

Passive wastewaters treatment consists of wetlands, limestone drains, and reactive barriers. Various passive methods are applied for the AMD treatment: natural or technogenic beds (these are cheap in construction and do not require substantial consumption of water); suppression of bacteria in waters by creating conditions the bacteria are not able to survive in (for example, increase of pH); prevention of rainwater ingress in order to avoid pyrite oxidation; addition of lime (lime is low-cost reagent and its application leads to decrease in sludge formation, but it is not widely used, because carbon dioxide forms buffers with pH below 6, while sometimes pH increase is crucial for some elements extraction, for example, Mn is usually extracted from solutions with pH far above 6); manufacturing of phosphate and nitrate fertilizers from the wastes by cation-exchange methods (reagents for this methods are quite expensive, but fertilizers are obtained as the output); and construction of wetlands (very effective treatment method as many heavy metals transform to insoluble compounds in these artificial wetlands) (Akcil and Koldas 2006; Sheoran and Sheoran 2006; Sivulka et al. 2007). Neutralization by limestone is the most environmentally justified. Though, this strategy is not always effective, because the limestone particles rapidly become covered with iron. Biological methods are cheap and safe, but before 2001 these methods were not well investigated. Only 20 % of companies have used biological methods by this time. Though biological methods have distinct advantages, they also are not free from disadvantages. For instance, some algae use AMD products for its life and growth. Acidophilic bacteria can be used for treatment of cyanides, as cyanides are essential for their life cycle. However, selected parameters (temperature and pH) and conditions such as composition of the medium should be maintained with care. From the wastes of granulated slag building materials can be obtained (Sivulka et al. 2007). Various methods for the treatment of wastewater from cyanides are used, such as clay, geomembrane from polyethylene, composite materials lining, providing a "dual control in the event of leakage" which is the best but the most expensive way, mainly used only by large companies.

The passive methods of minewaters treatment are much more cheaper than the active ones. However, they have several drawbacks limiting their use in the modern mining industries. First of all, some metals cannot be removed with these systems, for example Zn (Wolkersdorfer 2008). Second, they are usually less effective than the methods of active treatment. Moreover, the process of water treatment using passive methods is a long process, so this method cannot cope with the influx of wastewater during intense exploitation on multiple mines.

Process of minewaters purification is complex and includes different phases. Compared to other methods of wastewater purification, adsorption is one of the cheapest and therefore widely applied for removal of various types of pollutants (anions, metals, arsenic, mercury, radioactive elements, etc.). Nevertheless, adsorption cannot replace other methods fully, but may be used as alternative for some more expensive methods of minewater treatment. Therefore, in this study, adsorption considered as one of the methods in the overall chain of wastewater purification.

Searching of effective low-cost adsorbent is a focus of numerous research groups throughout the world. For example, a wide range of various low-cost adsorbents for different pollutants treatment is described in detail in Ali (2010) and Bhatnagar and Jain (2005). The following chapters discussed widely used commercial adsorbents, as well as cheaper modern alternatives.

Adsorption for mining wastewaters treatment

Innumerable physical, chemical, and biological processes take place at the boundary between two phases, while others are initiated at that interface. The change in concentration of a given substance at the interface as compared with the neighboring phases is referred to as adsorption.

Adsorption was first observed by C.W.Scheele in 1773 for gases, and followed by Lowitz's experiments in 1785 for solutions. There are different types of adsorption such as physical and chemical, from gases and liquids, with different adsorbents. In this review the adsorption of pollutants from mine industries wastewaters using low-cost adsorbents will be presented.

Wastewater purification by adsorption is carried on in two ways so called column method and batch method. Overall majority of researchers use batch method to study adsorption process, its parameters, and optimize conditions. Obtained results then are used for wastewater purification in industrial plants. These plants normally operate applying column adsorption method. Most researchers investigating new adsorbents for particular purpose use only one method in their works. Discussing pros and contras of any specific adsorbents as industrial one we need to take into account fundamental difference between these two approaches. As sometimes optimal conditions and suitability of particular adsorbent tested by batch method might be different from them when column method is applied (Ahmaruzzaman 2011a).

The choice of adsorption isotherms and kinetic models

The fundamental concept of adsorption science is adsorption isotherms describing equilibrium of material sorption at

Table 1 Adsorption isotherm for one- and poly-component systems

Name of isotherm	Equation	References
Henry's law	$q_e = Kq_mC$	(Dąbrowski 2001)
Langmuir	$q_e = \frac{Kq_mC}{1+KC}$	(Dąbrowski 2001)
Freundlich	$q_e = AC^B$	(Appel 1973; Liu 2005)
Brunauer–Emmett–Teller (BET theory)	$q_e = \frac{Kq_eC_r}{[1+(K-1)C_r][1-C_r]}$	(Maciej (2009)Caurie 2006)
Brunauer–Deming–Deming–Tellet (BDDT theory)	$q_e = \frac{Kq_{mCr}\left[1+m(g-1)C_r^{m-1}+gC_r^{m-1}(2g-1)C_r^m\right]}{(1-C_r)\left(1+[K-1]C_r+K(g-1)C_r^m-gC_r^{m-1}\right)}$	(Caurie 2006)
Redlich–Peterson	$q_e = \frac{Kq_mC}{(1+KC^B)}$	(Wu et al. 2010)
Langmuir–Freundlich	$q_e = \frac{Aq_mC^B}{1+AC^B}$	(Azizian et al. 2007)
Sips	$q_e = q_m\left[\frac{AC}{1+AC}\right]^B$	(Ahmed 2012)
Toth	$q_e = \frac{KC}{\left(1+\frac{C^B}{A}\right)+\left(1+\frac{C^B}{A}\right)^{\frac{1}{B}}}$, where $n_M = KA^{\frac{1}{B}}$	(Terzyk et al. 2003)
Yoon–Nelson	$\frac{C_e}{C_o} = \frac{1}{1+\exp[k_{YN}(r-(V/Q))]}$	(Yazıcı et al. 2009)
Dubinin–Radushkevich	$q_e = q_m \exp*\left\{2\left[\left(\frac{k_oe}{b_o}\right)^2\right]\right\}$	(Misra 1969; Condon 2000)
Dubinin–Astakhov	$q_e = q_m \exp*\left\{-\left[\left(\frac{k_oe}{b_o}\right)^2\right]\right\}$	(Jaroniec and Marczewski 1984; Stoeckli 1981; Condon 2000)
Temkin	$q_\mathrm{e} = \frac{R_\mathrm{g}T}{b_\mathrm{T}}\ln(A_\mathrm{T}C_\mathrm{e})$	(Khan 2012)
Fritz–Schlunder	$q_\mathrm{e} = \frac{q_\mathrm{m}K_\mathrm{FS}{C_\mathrm{e}}^{n_\mathrm{FS}}}{1+K_\mathrm{FS}{C_\mathrm{e}}^{m_\mathrm{FS}}}$	(Jossens et al. 1978)
BiLangmuir	$q_\mathrm{e} = \frac{q_{\mathrm{m1}}K_{\mathrm{BiL1}}C_\mathrm{e}}{1+K_{\mathrm{BiL1}}C_\mathrm{e}} + \frac{q_{\mathrm{m2}}K_{\mathrm{BiL2}}C_\mathrm{e}}{1+K_{\mathrm{BiL2}}C_\mathrm{e}}$	(Yamamoto et al. 1993)

a surface at constant temperature (Dąbrowski 2001). They allow to evaluate adsorption parameters, such as saturation rate of the adsorbent, the quality of the adsorbent, its selectivity, etc. The adsorption isotherm is an equation relating the amount of solute adsorbed onto the solid and the equilibrium concentration of the solute in a solution at a given temperature. The adsorption capacity of adsorbent is determined by use of adsorption isotherm. Isotherms used in wastewater treatment (Langmuir, Langmuir–Frendlich, Toth, Sips etc.) are presented in Table 1. The integral adsorption equation gives an opportunity to determine the mutual inter-dependence between the shape of the overall adsorption isotherm and the energy distribution function (Dąbrowski 2001).

Most often used adsorption isotherms are presented in Table 1. Suitable equations are chosen depending on process conditions. Unfortunately, there are no clear rules for correct and quick selection of the desired equation. The choice of a particular equation depends on many factors: composition of the system (one-, two-, or multi-component), properties of the adsorbent (single- or multi-layer adsorption), and the type of adsorption (chemisorption, physical adsorption, ion-exchange, or biosorption). The simplest approach is that at equilibrium one can assume that adsorption is proportional to concentration of adsorbed substance in the liquid phase. In this case, Henry's law is used to calculate the adsorption constant. The Langmuir isotherm assumes that adsorption occurs at specific homogeneous sites within the adsorbent without any interaction between the adsorbed substances. If data do not fit properly by linear equation, which is quite a common case, it is worth to try Freundlich isotherm. This isotherm is widely used despite it is only rough approximation from thermodynamical point of view. Isotherms BET and BDDT describe the process more precisely if filling of the adsorbent's pores with several layers of adsorbate takes place. Reduced concentration approach is used in these two isotherms, giving coverage parameter equal to one as far as pores are filled. BET isotherms are mainly used to evaluate the surface area of the adsorbent, and not for technological purposes. BDDT isotherm is particularly advantageous, since it explicitly take into account the number of adsorbed layers m and parameter of adsorption heat g.

In the equations of Redlich–Peterson, Langmuir–Freundlich, Sips, and Toth and Julian, new parameters were added to the Langmuir isotherm taking into account various deviations from linearity and the exponential mathematical form. Julian and Toth isotherm has a special purpose: it is an equation directly related to the data directory (Valenzuela and Myers 1989). This handbook is a great summary of pure components and mixtures, gases and liquids. There are other reference books containing information on adsorption isotherms (Dobbs and Cohen 1980; Yaws et al. 1995). Dubinin–Astakhov isotherm allows changing the exponent. In the equation of isotherm Dubinin–Stoekli, consider not just a specific pore size, but pore distribution in two sizes, which is a feature of many adsorbents. This equation can be expanded taking into account distribution of even more pore size numbers, although in practice it is applicable, taking into account maximum of three sizes. The Dubinin et al. isotherm equations in the various forms are used extensively to analyze porosity materials. These equations are no doubt widely used for the analytical forms for adsorption isotherms for different porous materials (Condon 2000).

In the industry the time dependence of adsorption on solid surfaces is named adsorption kinetics (Dąbrowski 2001). The theory of adsorption is well based on the kinetic processes occurring during adsorption. Various kinetic models help us to estimate the rate of adsorption as a function of the substances concentration and solutions pH (non-linear and linear pseudo-first- and second-order models (Lin and Wang 2009), intraparticle diffusion model (Wu et al. 2009), and Elovich equation (Cheung et al. 2000).

The effect of pH for kinetic process on chromium biosorption onto acorn shell adsorbent was studied by Aranda-García et al. (2010). Kinetic models satisfactorily describing chromium extraction from solution were Elovich and pseudo-second models, respectively. Freni et al. (2012) studied sorption effect for one, two, and four layers of loose silica grains. It was found out that another crucial parameter for dynamic behavior of the adsorption/desorption process is the size of adsorbent grains, as the process is much faster for smaller grains. Some authors received an erratic result, that poly-components systems are described by adsorption isotherms for mono-component systems (Kumar et al. 2008).

In systems with intraparticle diffusion being the rate-limiting step, the intraparticle diffusion approach is described by many scientists (Bridelli and Creppa 2008; Djekic 2007; McKAy et al. 1987; Ofomaja 2010; Papathanasiou and Bijeljic 1998; Rudzinski and Plazinski 2009).

Adsorbents

In this chapter, the materials and their adsorption properties, which are used as adsorbents for mining wastewater purification, or which may be of potential interest as adsorbents for these kinds of industrial wastewaters (with higher acidity and a large number of metal ions, cations, radioactive substances) are described.

Activated carbon

Carbons are the oldest and most widely used adsorbents. They have high surface area and developed porosity especially after activation process. Activated carbon is carbon produced from carbonaceous source materials such as nutshells, peat, wood, coir, lignite, coal, and petroleum pitch. It can be produced by physical or chemical activation. Comprehensive review of activated carbon from inexpensive materials is presented in Ali (2010).

Physical reactivation is precursor developed into activated carbons using gases. This is generally done by using one or a combination of the following processes, carbonization and oxidation processes. During the carbonization process, when material containing carbon is pyrolyzed at temperatures 600–900 °C in the absence of oxygen (usually in inert atmosphere with gases like argon or nitrogen). The oxidation process raw material or carbonized material is exposed to oxidizing atmospheres (carbon dioxide, oxygen, or steam) at temperatures above 250 °C, usually in the temperature range of 600–1,200 °C.

Chemical activation is prior to carbonization; the raw material is impregnated with certain chemicals. The chemicals are typically acids, strong bases, or salts (phosphoric acid, potassium hydroxide, sodium hydroxide, calcium chloride, and zinc chloride 25 %). Then, the raw material is carbonized at lower temperatures (450–900 °C). Chemical activation is preferred over physical activation due to lower temperatures and shorter time needed for activating material. The cost of activated carbon is about 500–1,800 USD per metric ton (Marsh and Reinoso 2006).

Granular activated carbons have been widely applied in wastewater treatment. Most carbonaceous materials are porous and have an internal surface area of around 10 m^2g^{-1}. One of the drawbacks preventing widespread use of activated carbon is its high price. However, the interest in activated carbons derived from a variety of materials such as peat, lignite, coconut shell, hardwoods, and other carbon-containing materials increased again (Acharya et al. 2009a; Acharya et al. 2009b; Dash et al. 2009; Deveci et al. 2006; Gupta et al. 2012a; Kadirvelu et al. 2001; McKay 1996; Sahu et al. 2009; Singh et al. 2008; Yeddou et al. 2010).

Bio-carbon adsorbents derived from forests biomass can be used in the wastewater treatment as well. Shin et al. (2008) observed outperformed adsorption properties of the samples produced from biomass than of commercial activated carbon from bio-mince for six metals (Cu, Cd, Mn, Ni, Pb, and Zn) extracted from AMD wastewaters. These adsorbents are suitable for AMD wastewaters; unfortunately, their use is limited by area where this type of forest can be found. Data are presented in Table 2.

Lignite, often referred to as brown coal, could be used as adsorbent for AMD treatment and removal of heavy metals from wastewater (Mohan and Chander 2006). The sorption capacity of lignite was found superior compared to commercial activated carbons in addition to its approximately two times lower cost (Table 2).

Yazıcı et al. (2009) studied cyanides treatment by oxidation and adsorption on activated carbon, nut shell, and rice husk from gold mining wastewater. It was shown that adsorption capacity of activated carbon was enhanced via impregnation with metals such as copper and silver (Table 2). Although activated carbons and agricultural by-products have relatively low capacity for cyanides removal they can be used (Adams 1994) to treat waters with low cyanide concentration (Table 2).

Activated alumina

Activated alumina (Spedding 1970) is manufactured from aluminium hydroxide by dehydroxylating it in a way that produces a highly porous material; this material can have surface area significantly over 200 m^2g^{-1}. The compound is used as a desiccant and as a filter for fluoride, arsenic, and selenium in drinking water. It is made of aluminium oxide (alumina; Al_2O_3), the same chemical substance as sapphire and rubies. Activated alumina is also widely used to remove fluoride from drinking water. In the US, there are widespread programs to fluoridate drinking water. However, in certain regions, such as the Jaipur region of India, there is enough fluoride in the water to cause fluorosis. Activated alumina filters can easily reduce fluoride levels from 0.5 ppm to less than 0.1 ppm. The amount of fluoride leached from the water being filtered depends on how long the water is being in contact with the alumina filter media. Basically, the more alumina in the filter, the less fluoride will be in the final, filtered water. Lower temperature water, and lower pH water (acidic water) are filtered more effectively too. The cost of activated alumina is approximately US $700–800 per metric ton which is about the same as activated carbon (Kapadia 2000). As shown in several recent studies (Ghosh and Gupta 2012; Lin and Wu 2001; Mor et al. 2007; Su et al. 2008; Wei et al. 2005), the modified alumina may be a good adsorbent for wastewater purification from variety of pollutants. This material cannot be classified at the moment as a low-cost adsorbent, as its modification increases the final cost substantially. However, if a cheap method of alumina modification is revealed the adsorbent will be of great potential for wastewater treatment in mining.

Zeolite

Among all the inorganic ion-exchange adsorbents zeolites have the best properties (Chester and Derouane 2010). The cost of zeolite is US $400–500 per metric ton. They have large surface area and high affinity to many metal ions (such as Pb, Cu, Cd, Zn, and UO_2). However, modified zeolites exhibit better adsorption properties than unmodified (Chester and Derouane 2010; Han et al. 2006; Motsi et al. 2009; Stanic et al. 2009; Vadapalli 2010; Zou et al. 2009), but modification

Table 2 Data on pollutants adsorption by different materials for mining wastewater purification

Sorbent	Pollutant	Pollutant's concentration, mgL^{-1}	pH	Temperature (°C)	Time, h	Adsorbent dosage (gL^{-1})	Adsorbent capacity (mgg^{-1})	Removal (%)	Adsorption isotherms	References
Activated carbon										
Activated bio-carbon	Cu	0.0005	2	–	–	10	720	–	BET	(Shin et al. 2008)
	Cd	0.004								
	Mn	0.97								
	Ni	0.012								
	Zn	0.32								
Activated carbon	$CN^{\frac{1-n}{n}}$	76	13	–	80	5	3.5–16.4	–	–	(Yazıcı et al. 2009
Activated carbon	NaCN	50	9	–	42	20	–	67	–	(Adams 1994)
	ZnCN		7					84		
	FeCN		5					82		
Lignite	Fe(II)	100	4	22	48	6	3.4	–	–	(Mohan and Chander 2006)
	Mn	100								
	Fe(III)	50								
	Zn	50								
Zeolite										
Zeolite	Fe^{3+}	400	2.5	22	6	30	6.41 mg/g	99.64	Langmuir and Freundlich	(Motsi et al. 2009)
	Cu^{2+}	20	3.5				0.54	99.99		
	Mn^{2+}	20					0.52	99.78		
	Zn^{2+}	120					2.21	96.34		
Fe(III)-modified natural zeolite tuff	As(V)	2	6–7	22.2	0.5	10	1.55 mg/g	99.00	Langmuir–Freundlich	(Stanic et al. 2009)
Zeolite-P from coal fly ash	Fe	104±3.5	8–9	–	–	10	–	99.9	BET (69 m^2/g)	(Vadapalli 2010)
	Al	181±1.2						77		
	Mn	20.5±2						64		
	Zn	284±2.6	7					69.2		
	Cu	18±3						68.6		
	Ni	701±25						88.9		
	Se	6±0.9						61.0		
Manganese-oxide coated-zeolite	U	100	4	20	3	5	15.1	99.90	Thomson	(Han et al. 2006)
Natural zeolite clinoptilolita	Pb	300–700	5		24	18	200.54	99	Langmuir	(Kleinubing and Silva 2008)
Chitosan										
Chitosan with 3,4-dihydroxybenzoi acid	U	10^{-2}	3	25	By column, $t1/2=$ 4 min	20	330	98.5	–	(Sabarudin et al. 2007)

Table 2 (continued)

Sorbent	Pollutant	Pollutant's concentration, mgL^{-1}	pH	Temperature (°C)	Time, h	Adsorbent dosage (gL^{-1})	Adsorbent capacity (mgg^{-1})	Removal (%)	Adsorption isotherms	References
Chitosan-tripolyphosphate beads	U	2,000	5	22	10	20	236.9	99.7	Langmuir	(Sureshkumar et al. 2010)
Chitosan	–	–	–	–	–	–	–	–	Langmuir	(Laus et al. 2007)
(AMD)	Fe	112	2.58	25	24	0.5	1.24	99.0	–	–
	Al	66						99.0		
	Cu	0.2	2.34					98.0		
(Decantation pooli)	Fe	446		25	24	2	0.562	99.0	–	–
	Al	136						98.0		
	Cu	06						95.0		
Chitosan	As	10	3.5	24±2	96	0.5	14.16		Langmuir Freundlich	(Kwok 2010)
Chiosan	As	11	5	24±2	48	0.5	2.5	92.3	Langmuir Freundlich	(Kwok 2009)
Crab-shell chitin	Al	16.2	–	–	480	4	–	>90.0	–	(Robinson-Lora and Brennan 2009)
	Fe	12.0								
	Mn	1.6								
	SO_4^{2-}	300								
Chitin complex (10 % chitin, 12 % protein, 78 % minera matter (35 % CaO))	AL	1,000	3–6.5 (in 3 days)	20	1,200	4	–	100	–	(Robinson-Lora and Brennan 2010)
	Fe							93		
	Mn							>73		
	SO_4^{2-}							98.22		
DTPA-modified chitosan	Co	100	2.1	22	–	2	49.1	–	Langmuir	(Repo et al. 2010)
	Ni						53.1		Freundlich	
Chitin products										(Pinto et al. 2011)
SC-80	Pb	1.1	6–7	22	24	2	1.24	>98		
	Fe							~100		
	Zn							~100		
	Cd	1.3	5–7				0.47–1.81	83–91		
							0.25–0.93			
	Co	0.78	7				0.98	37–59		
	Mn	52	6–7					64		
SC-20	Cd		6.48		120			87–93		
	Co							37–41		
	Mn							30–45		
Chitosan	Hg	50	4	25±2	2	5	109.55	98.0	Langmuir	(Benavente et al. 2011)
	Cu	50	6				79.94	97.8	Freundlich	

Table 2 (continued)

Sorbent	Pollutant	Pollutant's concentration, mgL^{-1}	pH	Temperature (°C)	Time, h	Adsorbent dosage (gL^{-1})	Adsorbent capacity ($mg g^{-1}$)	Removal (%)	Adsorption isotherms	References
	Pb	25					58.71	>97.0	Redlich–Peterson	
	Zn	50					47.15	71.3	Sips	
EDTA-modified chitosan	Co Ni	100	2.1	22		2	63.0 71.0		BiLangmuir Sips	(Repo et al. 2011a)
EDTA-modified chitosan-silica hybrid materials	Co Ni	(0.02–7 mM)	<3	22±1	24	2	(0.25–0.63)	93.5 95.2	BiLangmuir	(Repo et al. 2011c)
	Cd							96.5		
	Pb							99.2		
Silica gels										
DTPA-modified silica gel and chitosan	Co	10	3	22±1	12	2	–	93.3–93.8	BiLangmuir	(Repo et al. 2010)
EDTA- and DTA-modified silica gel	Co Ni	10	3.5	22±1	4	2	20 21.6	96–99	BiLangmuir	(Repo et al. 2009)
	Co						16.1			
	Ni						16.7			
Silica gel	SO_2	1,610	–	190	2	4.22	239	–	Freundlich Langmuir	(Kopaç and Kocabaş 2002)
									BET	
									Dubini–Radushkevich	
Modified silica gel	Co Ni	1–300	3	22±1	50	2	10	–	BiLangmuir Redlich–Peterson	(Motsi et al. 2009)
Biosorbents										
Dries green macroalga	Cu Cd	–	5	20±1	0.5	16.5	–	–	–	(Apiratikul and Pavasant 2006)
	Pb									
Sargassum filipendula	U	1.34	4	22	10	96.5 g	560	94.8	Langmuir Freundlich	(Rodrigues Silva 2009)
Arthrobacter	U	840 μM	5.8	25	1	1.5	2.580 μmol/g	100	Langmuir	(Tsuruta 2004)
Lactobacillus	U	840 μM	5.8	25	1	1.5	2.370 μmol/g	99.5	Langmuir	(Tsuruta 2004)
Bacillus	U	840 μM	5.8	25	1	1.5	2.500 μmol/g	95.5	Langmuir	(Tsuruta 2011)
Dairy manure composte	Pb Cu Zn	104 32 33	3.5 4.5 5.5	25±2	4	15	0.460 0.428 0.237	96.30 86.77 62.03	Langmuir Freundlich	(Zhang 2011)
Cupressus lusitanica Bark	Cr(VI) Cr(III)	800 800	5	27–29	168	1	87.5	–	Elovich	(Netzahuatl-Munor et al. 2012)

Table 2 (continued)

Sorbent	Pollutant	Pollutant's concentration, mgL^{-1}	pH	Temperature (°C)	Time, h	Adsorbent dosage (gL^{-1})	Adsorbent capacity ($mg\,g^{-1}$)	Removal (%)	Adsorption isotherms	References
Bacterium	As(V)	201.7	7.2		19 d	4.5	0.483	–	–	(Herbel and Fendorf 2006)
	As(III)	318.2					1.206			
Guar gum nitrilotriacetic acid	Fe(II)	1.03	5.5	25	4	1	–	96.37	–	(Singh 2012)
	Zn(II)	3.16						93.05		
	Cu(II)	0.76						88.67		
	Cd(II)	0.16						85.05		
	Pb(II)	0.66						74.25		
Municipal sewage sludge										
Municipal sewage sludge	Cu	48	6	–	–	10	17.3	99.9	Langmuir	(Phuengprasop et al. 2011)
	Cd	30	7				14.7	96.7		
	Ni	20								
	Pb	99								
			5				7.8	82.2		
							42.4	100		
Mine waste										
Iron slag	Cu	200	4.8	18	24	2	88.5	99.41	Langmuir	(Feng et al. 2004)
	Pb	200					95.24	99.94		
Steel slag	Cu	200	3.2	18	24	2	16.21	92.37	Langmuir	(Feng et al. 2004)
	Pb	200					32.26	96.23		
Waste mud from Cu-Zn industry	Pb	207	4	25	4	10	24.4	99.40	Langmuir Freundlich	(Ozdes et al. 2009)
Vanadium mine tailing	Pb	200	5.2	25	3	20	3.816	95.3	Freudlich	(Shi et al. 2009)
	Cr						3.868	99.1		
	Cu						3.240	91.2		
	Cd						2.844	94.9		
	Ni						2.207	98.0		
Iron ore slims	Pb	20–500	5.1	28	5–270	10		95	Langmuir	(Panda et al. 2011)
	Cd							80	Freundlich	
	Cu							70		
Industrial waste sludge	Zn	5,000	5	25	3	20	7.26	88.00	Langmuir	(Umesh 2012)
									Freundlich	
									Redlich–Peterson	
									Tempkin	
Red mud	Cu	100	6	–	4	10	–	98.03	–	(Çoruh S and Ergun 2011)

Table 2 (continued)

Sorbent	Pollutant	Pollutant's concentration, mgL^{-1}	pH	Temperature (°C)	Time, h	Adsorbent dosage (gL^{-1})	Adsorbent capacity ($mg g^{-1}$)	Removal (%)	Adsorption isotherms	References
Comparative analysis of other adsorbents										
Peat fly ash	Zn	23.5	6.3	–	12 h	–	21.4	99.9	–	(Warrender 2011)
	Pb	0.5					0.88	98.6		
	Cd	0.05					0.04	99.8		
Compost	Zn	23.5					2.48	99.7		
	Pb	0.5					0.74	99.2		
	Cd	0.05					0.014	95.2		
Waste shell material	Zn	23.5					1.72	79.9		
	Pb	0.5					0.63	98.1		
	Cd	0.05					0.007	80.6		
BOS/BFS slag	Zn	23.5					0.20	56.8		
	Pb	0.5					0.08	99.9		
	Cd	0.05					0.0015	76.0		
Crop residues	Fe	2-, 5-, 10-, and 25-fold from the 0.1 M concentration solution	7	70	4	20	0.84	82.0	Langmuir	(Mosa et al. 2011)
	Mn						21.48	48.0		
	Zn						20.03	75.0		
	Co						618.99	69.0		
	Cu						11.38	83.0		
	Pb						0.51	99.9		
	Ni						14.62	70.0		
	Cd						468.84	77.0		
Dairy manure compost	Pb	0.5	4	25±2	1	15	0.460	97.4	Langmuir	(Zhang 2011)
	Cu						0.428	96.5	Freundlich	
	Zn						0.237	94.1		

process increases the cost of treatment. One should keep in mind that the adsorption capacity of zeolite declines with temperature increase, therefore, modification is recommended to be carried out at low temperatures (Motsi et al. 2009; Vadapalli 2010). Motsi et al. (2009) conducted comparative analysis of adsorption of components mixture (Fe^{3+}, Zn^{2+}, Cu^{2+}, and Mn^{2+}) and individually for each component with synthetic AMD using natural zeolite. It was found that presence of competing ions did not affect iron adsorption. Natural zeolites adsorbed copper and manganese 30 % more effective than synthetic one (Table 2). Vadapalli (2010) studied the zeolite from AMD with high cation activity and surface area. The data describing multiple use of the material were also presented. It was reported that for such metal ions as Al^{3+}, Fe^{3+}, Mn^{2+}, Zn^{2+}, Cu^{2+}, and Ni^{2+}, the high degree of purification was reached during the multiple re-use of material. For such ions as K^{+}, Ca^{2+}, Sr^{2+}, and Ba^{2+}, studied zeolite was effective only during the first use of material (Table 2). Zeolite modified with iron(III) has better adsorption properties for arsenic(V), than non-modified natural zeolite (Stanic et al. 2009). Stanic et al. (2009) fitted adsorption data for various isotherm models, such as Langmuir, Freundlich, and Langmuir–Freundlich. Langmuir–Freundlich model having r_2 value of 0.99 and the best fit for the experimental adsorption data (Table 2). Along with the study of Cu(II) and Pb(II) treatment from wastewater (Han et al. 2006), authors was studied uranium treatment (Table 2). A new adsorbent of manganese-coated zeolite was synthesized and its adsorption properties for uranium removal have been studied (Zou et al. 2009). This work established that the new material is an effective adsorbent for uranium(VI) extractions from wastewaters. However, the presence of competing ions (Zn^{2+} or Cu^{2+}) in solution reduces the adsorption of uranium. Natural zeolite showed very high adsorption capacities for Pb(II) removal from water in batch systems (Kleinubing and Silva 2008). Data are shown in Table 2.

Chitosan

Chitosan is a material for the preparation of adsorbents, which is widely used for wastewaters treatment. The use of crab-shell chitin may be the most economically proven solution at the field scale, since it is considerably cheaper on a mass-basis than other currently available substrates (US $0.20–0.60 per metric ton for crab-shell chitin vs. $50 per metric for spent mushroom compost and US $20–30 per metric for limestone) (Robinson-Lora and Brennan 2010). It is used in modified form in many methods. Summarized data on the modified chitosan are presented below in Table 2.

3,4-dihydroxybenzoic acid-modified chitosan has shown good adsorption properties for the adsorption of 60 elements from river and seawaters. Better selective adsorption of uranium(VI) (adsorption capacity was 330 $mg g^{-1}$) was observed of acid modified chitosan compared to non modified and modified by other methods (Sabarudin et al. 2007) (Table 2). Although the experiments were carried out for natural waters, this acid modified chitosan might be of interest for adsorption from wastewater.

Oshita et al. (2009) have modified cross-linked chitosan by catechol, iminodimetylphosphonic acid, iminodiacetic acid, phenylarsonic acid, and serine and have studied their properties for adsorption of uranium(VI) and others elements (such as Be(II), Mn(II), Co(II), Ni(II), Cu(II), Zn(II), Ga(III), Y(III), Ag(I), Cd(II), In(III), Pb(II), Bi(III), and Th(IV)) from wastewater.

The cross-linked chitosan modified with catechol was suitable for the selective and effective sorption of uranium. The data obtained in the study (Sureshkumar et al. 2010) of uranium adsorption by chitosan modified with tripolyphosphate showed that this modification decreased adsorption capacity (236.9 $mg g^{-1}$) as compared to other modification methods (Sabarudin et al. 2007).

The properties of metal (Fe(II), Al(III), and Cu(II)) adsorption by chitosan from various real coal mining wastewaters were observed (Laus et al. 2007). It was shown that this material was a good adsorbent for the treatment of AMD. In the adsorption process of the “decantation pool” material properties deteriorated, perhaps it was due to sharp increase in metal concentration in the sample (Table 2), and the fact that the adsorbent was used four times. It is likely that the matrix solution interfered the adsorption of metals and that competitive additional influence on the adsorption process of the studied metals occurred.

Adsorption of arsenic(V) from wastewater is easier than arsenic(III) (Kwok 2010; Kwok 2009). Conversion of As(III) to As(V) by pH maintaining can solve this problem. According to these studies, we can conclude that more research is required as the adsorption of arsenic by chitosan is still poorly investigated. In both studies the adsorption of arsenic at pH equal to 4 was approximately 96 % (Table 2).

Interesting results were obtained with the use of crab-shell chitin (Robinson-Lora and Brennan 2009) and chitin complex (Robinson-Lora and Brennan 2010) in AMD: effective sulfate removal followed by pH raising to near neutral in less than 3 days, facilitates efficient removal of dissolved metals (Fe(III), Mn(II), and Al(III)). Double column and microcosm test were examined to AMD treatment. The microcosm test was more effective for these conditions with the use of crab-shell chitin as an adsorbent (Robinson-Lora and Brennan 2009; Robinson-Lora and Brennan 2010). The high efficiency of chitin complex in removing Mn(II) in comparison to the other evaluated substrates makes it a very attractive alternative for sites with high levels of this metal (Robinson-Lora and Brennan 2010). Data are presented in Table 2.

Pinto et al. (2011) investigated the adsorption properties of two different materials SC-20 (raw crushed crab shells containing 40 % CaCO3, 30 % protein, 20 % chitin, 7 % moisture, and 3 % ash) and SC-80 (the chitin polymer containing 88 % chitin and 12 % water) in different acidity conditions. It was found that SC-20 is much more effective than SC-80 for adsorption of Fe(III), Pb(II), Zn(II), Cd(II), Co(II), Cu(II), and Mn(II), thus pH control is needed. Pb(II), Mn(II), Cd(II), and Co(II) was partially removed only with SC-80 material. It could be due to the fact that the SC-20 contains $CaCO^3$, which effectively removes metals and neutralize acidity (Table 2). Benavente et al. (2011) showed that solution pH strongly affects the adsorption capacity of chitosan while adsorbing Cu(II), Hg(II), Pb(II), and Zn(II) from gold mining wastewaters. The equilibrium data were fitted by Langmuir, Freundlich, Redlich–Peterson, and Sips isotherm models for different elements. For example, the Redlich–Peterson isotherm ideally fits observed well of Hg(II) and Zn(II) removal, Langmuir and Sips isotherms are the best models for Cu(II) and Pb(II), respectively (Table 2).

Ethylenediaminetetraacetic acid (EDTA)-modified chitosan was found more effective for Co(II) and Ni(II) removal from liquids than diethylenetriaminepentaacetic acid (DTPA)-modified chitosan (Repo et al. 2010). Repo et al. (2011a, 2011c) also studied the properties of combined adsorbents made of EDTA-modified chitin and DTPA-modified silica gel. These adsorbents revealed good adsorption properties for Co removal, and EDTA-modified chitosan was effective for Co(II), Ni(II), Cd(II), and Pb(II) removal from the metals solution (Repo et al. 2011c) (Table 2).

Silica gel

Silica gels as adsorbents have been known for almost a century (Iljin 1932; Stuart and Coull 1958). It was used in World War I for the absorption of vapors and gases in gas mask canisters.

Silica gels adsorbed effectively vapors of many organic substances along with water (Gushikem and Da Silva 1985; Moriguchi et al. 2005; Parida et al. 2006). This property used for recovery of gasoline, benzene, ether, acetone, etc. vapors from air and natural gas (Amini et al. 2011). Kopaç and Kocabaş (2002) investigated adsorption of sulfur dioxide on silica gel (Table 2).

However, recently these materials attracted scientist's attention not only as adsorbent of various air pollutants, but as adsorbent from aqueous systems. This is due to its excellent adsorption characteristics, such as surface areas and polarity. For example, the surface area of silica gels ranges from 100 to 750 $m^2 g^{-1}$, despite the fact that the surface area of commercial activated carbons is 500 $m^2 g^{-1}$. This is a fairly high value and thus silica gels can be assumed to be good adsorbents for aqueous systems treatment. This assumption has been confirmed by a range of scientific works conducted in this area over the last couple of years. However, silica gels have one great demerit from the environmental point of view as they are not biodegradable in either water or soil (Freni et al. 2012; Liu et al. 2012; Mahmoud et al. 2011; Repo et al. 2011b; Repo et al. 2009; Taha et al. 2012; Wantala 2012).

Repo et al. (2011b, Repo et al. 2009) investigated Co(II) and Ni(II) removal from model liquids with silica gel materials functionalized with both EDTA and DTPA. The removal of Co(II) and Ni(II) by modified silica gels was substantially higher than that by their unmodified form (Table 2).

Taha et al. (2012) used silica gels as composite nanofibrous material for membranes preparation. These membranes showed good adsorption properties for Cr(VI) removal from aqueous solution. The maximum adsorption capacity was 19.46 $mg g^{-1}$ and adsorption behavior was described by the Langmuir adsorption isotherm.

Silicon dioxide nanopowder and *Aspergillus ustus* combination was studied by Mahmoud et al. (2011) as adsorbent material for Cd(II) removal from real aqueous samples. Maximum adsorption capacity was 1 $mol g^{-1}$ at pH 7. Sorption equilibria were established in 20 min, data were fitted by both Langmuir and Freundlich adsorption models.

Biosorbents

Materials such as agricultural residues, algae, and microbial species can be used as biosorbents. There are some reviews about agricultural residues as potential adsorbents for wastewaters treatment of organic compounds and toxic metals (Bhatnagar and Sillanpää 2010; Demirbas 2009; Dias et al. 2007; Mahmoud et al. 2011; Salleh et al. 2011; Smith et al. 2009b; Sud et al. 2008). As it was mentioned above in activated carbons chapter a variety of materials such as peat, lignite, coconut shell, hardwoods, and others can be treated forming effective sorbents. Agricultural by-products and their modified materials produced from them have shown high capacity for metal adsorption. This is due to the functional groups presenting in agricultural waste biomass such as acetamido, alcoholic, carbonyl, phenolic, amido, amino, and sulphydryl groups capable of forming complexes or chelates with heavy metals. However, all these materials need pretreatment or/and need to be modified to become effective adsorbents.

Tang et al. (2009) presented overview of bacteria role in sulfur cycle for mine wastewater. In this article a description of studies done during the last decade and earlier can be found. Biooxidation of sulfide and sulfur compounds is carried out in biotreatment of AMD. Often this method is used for passive wastewater treatment, which is less efficient than active methods as described above. However, AMD treatment with bacteria can be the subject of more detailed future studies.

As it was mentioned above, some microbial species and algae have high accumulating capacity for radioactive elements (Rodrigues Silva 2009; Tsuruta 2011; Tsuruta 2004), metals (Gupta et al. 2010; Jiménez-Rodríguez et al. 2009; Lakaniemi et al. 2010; Netzahuatl-Munor et al. 2012; Singh 2012; Zhang 2011), and arsenic (Herbel and Fendorf 2006); it allows to use them for pollutants removal from mine tailing and wastewaters. For example *Sargassum filipendula* removal (99.7 %) enriched uranium from wastewater in 10 h (Rodrigues Silva 2009). *S. filipendula* showed good results for the adsorption of uranium and other metals. The maximum uranium adsorption capacity was 560 $mg g^{-1}$ at pH 4, with the maximum concentration of uranium in the solution equal to 50 $mg L^{-1}$, however, saturation of the biomass was not reached. Freudlich model described uranium biosorption better than the Langmuir model (Table 2). The adsorption of metals such as Ba(II), Pb(II), and Cr(V) from the wastewater was observed along with the adsorption of uranium. However, the concentration of Ca(II), Mg(II), Fe(III), and Mn(II) was risen by 29, 31.3, 16.5, and 11 %, respectively. These elements are part of the algae's polysaccharides and it is most likely that ion-exchange process takes place. In the later work (Zhang 2011) the same effect was observed: release of Ca^{2+}, Mg^{2+}, K^{+}, and H^{+} as a result of adsorption of Pb^{2+}, Cu^{2+}, and Zn^{2+} in dairy manure compost (DMC; Table 2).

In the book, Tsuruta (2011) describes a study of uranium removal from waste- and seawater with different types of bacteria. It was found that *Bacillus* sp., *Arthrobacter* sp., and *Lactobacillus* sp. are the best choice for adsorption (Table 2) to dry microbial cells was approximately at 2,500 $\mu mol g^{-1}$ adsorption capacity from water at pH 7–5 uranium. *Arthrobacter* sp. showed good results for adsorption of UO^{22+}, Cu^{2+}, Cd^{2+}, Mn^{2+}, Zn^{2+}, Ni^{2+}, and Co^{2+} (1 h at 25 °C, pH 5.0; UO^{22+} Cu^{2+} others). The presence of uranium in solution does not affect the adsorption of thorium and in average more than 98 % of thorium is extracted, while the adsorption of uranium decreases almost twice in presence of thorium.

DMC of AMD treatment can be used only at high pH, because they have adsorption effectiveness at low pH (Netzahuatl-Munor et al. 2012). However, Zhang (2011) was describing adsorption of Pb^{2+}, Cu^{2+}, and Zn^{2+} on DMC from wastewater at pH=3 and adsorption effectiveness for these metals was 93, 77, and 47 %, respectively. Optimum pH for these heavy metals was 3.5, 4.5, and 5.5, respectively. The maximum adsorption capacities calculated by applying the Langmuir equation to single ion adsorption isotherm were 0.460 $mmol g^{-1}$ for Pb(II), 0.428 $mmol g^{-1}$ for Cu(II), and 0.237 $mmol g^{-1}$ for Zn(II) at pH 4.0. The single and competitive adsorption studies showed that the adsorption affinity order of the three heavy metals was Pb(II)>Cu(II)>Zn(II). These results suggest that the total adsorption capacity of heavy metals in multi-solute systems increases, but individual metal ion adsorption will decrease due to metal ions competition for adsorption sites. The regeneration experiments showed that the adsorbent could be regenerated and reused at least three cycles without significant decrease in adsorption capacity. The results showed that DMC could have great potential in treating heavy metals in acid mine drainage. Finally, ion exchange was found to be the primary mechanism of adsorption.

Singh (2012) prepared the guar gum nitrilotriacetic acid, where the starting material was guar gum, which is cheap and easily available as agricultural resource. Experiments in columns of water purification from toxic metals were carried out. Adsorption properties of reused materials were also studied. Adsorption capacity remained at the same level after three cycles of material use. It was found that the GNTAA resin is effective adsorbent for removal of different toxic metal ions from effluent, and follows the order Fe(II)>Zn(II)>Cu(II)>Cd(II)>Pb(II) (Table 2).

Municipal sewage sludge

Recently, municipal sewage sludge has been of increasing interest as a material of adsorbents for wastewater treatment due to high carbon concentration in biochemical sludge. The municipal sewage sludge can be considered as low-cost adsorbents with high surface area. The application of adsorbents from sewage sludge wastes for wastewater treatment from other pollutants, such as metal ions and organic compounds, has been studied (Gómez-Pacheco et al. 2012; Handojo 2009; Rozada et al. 2008). Sewage sludge wastes need modification before use. For example Phuengprasop et al. (2011) modified municipal sewage sludge by iron oxide and used them for metal ion removal. The adsorption of Cu(II), Cd(II), Ni(II), and Pb(II) was studied using batch method (Table 2). It was observed that the presence of $NaNO_3$, $Ca(NO_3)_2$, and Na_{24} in metal solution in the concentration of 0.01, 0.01, and 0.50 M could reduce the removal efficiency. In review Smith et al. (2009a) different methods of sewage sludge-based adsorbents activation were observed, such as carbonization, physical and chemical activation and these methods combinations. According to review Smith et al. (2009a) chemical activation utilizing alkali metal hydroxides is the most effective technique for producing high surface area adsorbents from sewage sludge wastes. In review by Bhatnagar and Sillanpää (2010) utilization of agro-industrial and municipal waste materials as potential adsorbents for wastewater treatment was fully considered, and the detailed table with adsorbents, adsorbates, and adsorption capacities was provided. These data allow us to conclude that the municipal sewage sludge is quite promising for wastewater treatment in the mining and metallurgical industries. However, at this stage of their study, they are unlikely to be of commercial interest because of their preparation and modification, increasing the final costs of treatment method substantially. Although, if cheaper methods of municipal sewage sludge pre-treatment are found, adsorbents of this type will be of great interest in mining wastewaters treatment. Since this approach solves several problems, first of all reduction of environmental risks in mine wastewater treatment by materials, which are wastes themselves.

Mine waste

One of the most advantageous ideas for modern industries is the idea of a waste-free production, when wastes are used for wastewater treatment on the same factories (Ahmaruzzaman 2010; Ahmaruzzaman 2011b; Ai-qin et al. 2009; Bhatnagar and Jain 2005; Feng et al. 2004; HE et al. 2010; Ozdes et al. 2009; Panda et al. 2011; Potgieter-Vermaak et al. 2006; Shi et al. 2009; Ulrich 1999; Umesh 2012; Çoruh and Ergun 2011; Gupta et al. 2012a; Gupta et al. 2012b; Gupta and Sharma 2003; Jain et al. 2004). It is likely impossible to find a common approach for the wastewater purification of complex composition. Therefore, different methods are combined, or a mixture of different adsorbents is used. It strongly depends on the composition of wastewater under purification, as well as treatment conditions. The combination of adsorption and following flotation has been proven as an effective method for removal of heavy metals from wastewater streams by Feng et al. (2004). Adsorption properties of iron and steel slag for Cu(II) and Pb(II) removal from wastewater were studied. Adsorption capacity was considerably higher for the iron slag compared to steel slag adsorbent. The difference is probably caused by higher surface area, porosity, and ion-exchange ability. Ion-exchange and sorption properties are well combined with redox capacity of the iron slag. The adsorbing slag flotation is particularly attractive for the removal of heavy metal ions from acid streams and promising results have been obtained on the treatment of an acid mine drainage.

Feng et al. (2004), Ozdes et al. (2009), Panda et al. (2011), and Shi et al. (2009) described different adsorbents used for mine wastewater purification from Pb(II), Cu(II), Cd(II), and Ni(II), as they are basic pollutants in mine industry. All adsorbents showed high affinity for lead ions (removal was approximately 99 %), other metals were adsorbed of 60 to 80 % (Table 2). The similar results for extraction ratio could be explained by the similarity of the adsorbent composition. Adsorbents were procured from mine industries waste, as iron ore slimes, metallurgical by-product slags, waste muds from copper mine industry, and vanadium mine tailing. All of them, except the last, have similar compositions (Table 3). It could be seen from the composition of adsorbents that an exchange is the basic process in wastewater purification; in particular, the exchange of the metal and calcium ions in the silicon oxide complex (Feng et al. 2004).

$$= \mathrm{SiOCa} + \mathrm{M}^{2+} \rightarrow = \mathrm{SiOM} + \mathrm{Ca}^{2+}$$

The mechanism of the process is similar to one described in the (Panda et al. 2011; Shi et al. 2009).

$$\begin{aligned} &= \beta\mathrm{S{-}OH} + \mathrm{M}^{n+} \leftrightarrow \equiv \mathrm{S{-}OM}^{(n-1)+} \\ &= \beta\mathrm{S{-}O}^{-} + \mathrm{M}^{n+} \leftrightarrow \equiv \mathrm{S{-}OM}^{(n-1)+} \end{aligned}$$

where ≡ S-OH denotes the aluminol or silanol groups on the surface of the mineral, *M* is the heavy metal ions. This mechanism explains why the adsorption is better at pH 3.5, as the systems equilibrium is moving to the right with increasing of pH. These adsorbents exhibit high affinity for the lead ions. The extraction ratios of other metals adsorbed from wastewater are substantially lower (Feng et al. 2004; Panda et al. 2011), sometimes lower than 50 % (Ozdes et al. 2009; Shi et al. 2009).

In addition to Pb(II), Cu(II), Cd(II), Ni(II), and Zn(II) other metals may occur in mining wastewaters. Removal of zinc ions from wastewater using industrial waste sludge was

Table 3 The composition of materials from industrial wastes

Iron ore slimes (Panda et al. 2011)		Metallurgical by-product slag (Feng et al. 2004)		Waste mud from copper mine industry (Ozdes et al. 2009)		Vanadium mine tailing (Shi et al. 2009)	
Fe_2O_3	65.50	CaO	46.30	Fe_2O_3	48.40	O	36.20
Al_2O_3	13.00	SiO_2	35.60	SiO_2	11.70	C	28.47
SiO_2	12.80	FeO	15.60	Al_2O_3	8.90	Si	21.10
		Al_2O_3	10.30	ZnO	0.70	Al	5.99
		MgO	5.90	CuO	0.70	K	2.48
		S	0.30	Other constituents		V	1.45
					29.60	Fe	1.09
						Mg	0.88

studied by Umesh (2012). The maximum adsorption capacity of Zn(II) by waste sludge was calculated as 7.26 $\mathrm{mg\,g^{-1}}$ which is fairly compared to other adsorbents (Table 2). The equilibrium data for Zn(II) removal were fitted by various adsorption isotherm models, but Langmuir isotherm model was found to be the most suitable. It should be taken into account that the paper described only Zn(II) removal, while normally wastewaters contain other elements able to reduce adsorption properties. In order to confirm with confidence that this adsorbent is suitable for wastewater treatment, additional research is required to study possible interference of other waste components.

Çoruh and Ergun (2011) studied Cu^{2+} removal from wastewaters by using red mud—waste from aluminium industry. Almost 90–98 % of copper adsorption from aqueous solution at initial concentration of 12.5–200 $\mathrm{mg\,L^{-1}}$ was observed. The results indicate that the red mud can be successfully used for the removal of Cu^{2+} in wide range of concentrations. Possible interference of other waste components was not studied.

Ahmaruzzaman (2011a) described wide range of industrial wastes as adsorbent for removal of heavy metals from wastewaters. This review provides a critical analysis of these industrial wastes, describes their characteristics, and also presents some of their drawbacks. It was noticed in this paper that adsorption in a column was better than in batch systems. However, there are authors who suggest otherwise (Johnson and Hallberg 2005; Kwok 2010). This may be due to the fact that more careful selection of the conditions is required for experiments in columns. If Ahmaruzzaman (2011a) hypothesis is correct, most probably it is due to continuous contact of the same concentration of adsorbate with investigated adsorbents in columns. Whereas in batch systems concentration of extracted compounds is steadily decrease, thereby decreasing the effectiveness of the adsorbent. However, in both cases one needs to take into account processes of adsorption and desorption. Equilibrium in batch systems is reached at a certain time, which can be easily determined experimentally. Therefore equilibrium needs to be controlled in the system to desorption. Most likely the choice of adsorption method should be considered separately in each case, based on the economic aspects.

Comparative analysis of other adsorbents

Some researchers have carried out comparative analysis of the adsorption properties of different structures and origin materials. For example Warrender (2011) studied the ability of five low-cost materials (compost, fly ash, waste shell material, iron ochre, and a mixture of blast furnace slag (BFS) and basic oxygen furnace slag (BOS)) to remove Zn(II), Pb(II), and Cd(II) from Fe-poor mine waters at neutral pH. The fly ash from peat was the most effective among others. However, all five materials showed good properties and are suitable of Pb(II) removal from wastewaters. Before use as adsorbents all these materials except fly ash require modification. According to many studies chemically modified materials exhibit higher adsorption capacities than unmodified forms. In addition "the application for example of untreated plant wastes as adsorbents can also bring several problems such as low adsorption capacity, high chemical oxygen demand, and biological chemical demand as well as total organic carbon due to release of soluble organic compounds contained in the plant materials" (Wan Ngah et al. 2011). Wan Ngah et al. (2011) reported the use of chemically modified plant wastes including rice husk, spent grain, sawdust, sugarcane bagasse, fruit wastes, weeds, and other as adsorbents for removal of heavy metal ions such as Cd(II), Cu(II), Pb(II), Zn(II), Ni(II), and Cr(VI) from wastewaters (Table 2). It was shown that chemically modified plant wastes vary greatly in their ability to adsorb heavy metal ions from wastewaters. For example *Alfalfa* biomass, rice husk, banana stem, and *Azola filiculoides* modified by sodium hydroxide, tartaric acid, formaldehyde, and hydrogen peroxide-magnesium, respectively have high adsorption effect the removal of lead, compared to materials such as bagasse fly ash modified with hydrogen peroxide, sawdust, and walnut sawdust modified formaldehyde in sulfuric acid. Adsorption capacity for modified sugarcane bagasse used for Pb removal varies greatly with modification methods. Mosa et al. (2011) studied efficiency of different crop residues and chemical pretreatment solutions for the removal of heavy metal ions (Fe(III), Mn(II), Zn(II), Co(II), Cu(II), Pb(II), Ni(II), and Cd(II)) at different concentrations from synthetic wastewater solutions. These materials were modified by different methods in order to enhance their adsorption properties. Cotton stalks were found the best adsorbent for metals removal from aqueous solutions among other materials. Lead is removed from the solution better than other metals using all the above adsorbents, whereas manganese removal was the most difficult. Zhang (2011) studied adsorption of Pb(II), Cu(II), and Zn(II) from simulated AMD using manure compost. It was shown that the presence of mixed heavy metal ions had little effect on sorption capacity of Pb, followed by Cu(II), with Zn(II) being most affected. Ríos et al. (2008) investigated coal fly ash, natural clinker, and synthetic zeolites (faujasite) as potential adsorbents for wastewater treatment. Their experiments showed that faujasite effectiveness as adsorbent is decreasing in a row Fe(III), As(V), Pb(II), Zn(II), Cu(II), Ni(II), and Cr(II), respectively. Recently, a lot of works about nanoparticles adsorbents were carried out. This modern trend will certainly have a great future when nanoparticles synthesis becomes cheaper (Ali 2012).

Conclusion

Finding adsorbents for wastewater purification is a question of high priority for mining industry. There is a wide range of requirements such as adsorption capacity, selectivity, kinetic properties, environmental impact, and the last but not the least the cost of adsorbent. The paper reviews properties of the cheapest available adsorbents suitable for wastewater treatment in mining industry: adsorbents made from municipal and industrial wastes, chitosans, and zeolites. Adsorbent choice depends heavily on wastewater composition.

Unfortunately at the moment there is no universal adsorbent for extraction of toxic metals, arsenic, mercury, sulfates, and cyanides. Therefore, most likely, researchers need to look for suitable combination of different adsorbents in each case.

It seems promising to use adsorbent modification to change their surface and therefore adsorption properties, sometimes modified materials demonstrate better adsorption properties than commercial analogues. Choosing modification method one needs to take into account properties of modified material, modification costs, environmental impact of the process.

There are limited data of the competitive adsorption of metals from liquids. Further researches are required in this area to optimize conditions for wastewater treatment and future application of the developed techniques in industries.

Since industries have to pay for wastes utilization it can be profitable to use their own wastes as a base material for adsorbent production especially taking into account the fact that some of their wastes show great potential for the purpose. In this case the possibility of contamination of tread water with by-products from the adsorbents should be kept in mind. One needs to take into account this pollutants "exchange" and consider pretreatment of wastes used as adsorbents in order to prevent possible pollution. Adsorbents production from industrial wastes is a good opportunity to solve several problems, such as waste utilization, wastewater purification, and as a result to cut costs.

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