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Bio-recycling of metals: Recycling of technical products using biological applications

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Abstract

The increasing demand of different essential metals as a consequence of the development of new technologies, especially in the so called “low carbon technologies” require the development of innovative technologies that enable an economic and environmentally friendly metal recovery from primary and secondary resources. There is serious concern that the demand of some critical elements might exceed the present supply within a few years, thus necessitating the development of novel strategies and technologies to meet the requirements of industry and society. Besides an improvement of exploitation and processing of ores, the more urgent issue of recycling of strategic metals has to be enforced. However, current recycling rates are very low due to the increasing complexity of products and the low content of certain critical elements, thus hindering an economic metal recovery. On the other hand, increasing environmental consciousness as well as limitations of classical methods require innovative recycling methodologies in order to enable a circular economy. Modern biotechnologies can contribute to solve some of the problems related to metal recycling. These approaches use natural properties of organisms, bio-compounds, and biomolecules to interact with minerals, materials, metals, or metal ions such as surface attachment, mineral dissolution, transformation, and metal complexation. Further, modern genetic approaches, e.g. realized by synthetic biology, enable the smart design of new chemicals. The article presents some recent developments in the fields of bioleaching, biosorption, bioreduction, and bioflotation, and their use for metal recovery from different waste materials. Currently only few of these developments are commercialized. Major limitations are high costs in comparison to conventional methods and low element selectivity. The article discusses future trends to overcome these barriers. Especially interdisciplinary approaches, the combination of different technologies, the inclusion of modern genetic methods, as well as the consideration of existing, yet unexplored natural resources will push innovations in these fields.

Keywords: *Biohydrometallurgy, Biomining, Bioleaching, Biosorption, Bioflotation, Bioreduction, Metal recovery, Recycling, Critical metals*

1. Introduction

Metals are important for all aspects of our daily life. Their increasing demand in the course of the last centuries put a permanent pressure on natural resources (UNEP, 2013). Due to society changes, a growing world population, and new technologies and future products, an increasing demand of metals can be expected.

The primary production and industrial processing of metals and widespread use in many products and industry resulted in the release of many metals to the environment (Bakhiyi et al., 2018; Robinson, 2009; Schüler et al., 2011; UNEP, 2013). The pollution with toxic metals is mainly caused by the release of the metals in effluent waters from mines, landfills, factories, and metal refineries. Further, current recycling rates are low for many technical products and rare metals. Recycling has become increasingly difficult due to the complexity of products and diverse interactions within recycling systems (Dodson et al., 2012; UNEP, 2013). Especially many modern electronic products contain small amounts of metals with a complex composition that complicate their recovery. For example, a modern car contains nearly all metals available. Despite valuable elements e-wastes contain many environmental organic or inorganic contaminants such as polychlorinated biphenyls, polycyclic aromatic hydrocarbons, antimony, chromium, nickel and others that potentially can be released to the environment (Bakhiyi et al., 2018; Robinson, 2009). Further, the promoted “low carbon technologies” that are considered as environmentally friendly such as wind turbines, solar cells, energy saving light bulbs, fuel cells and catalytic converters require rare and precious metals (Dodson et al., 2012; Jacobson and Delucchi, 2009). Currently the majority of these elements are mined and extracted from primary ore in highly energy intensive processes (Dodson et al., 2012; Hunt et al., 2013). Mining activities have a great impact on environmental conditions. They have led to elevated levels of metals in surrounding soil and water courses, resulting in destruction of vegetation and crops (Dodson et al., 2012; Schüler et al., 2011). Further, there are severe concerns regarding the availability of these elements in the future due to their low abundance and difficulty to access.

Recycling is an important approach to save resources. Production from primary resources consumes high amounts of energy, water, land, and chemicals, produces high amounts of wastes and has a serious impact on the environment (Schüler et al., 2011). Therefore especially rare metals should be reused as much as possible. In addition, an efficient recycling secure resource requirements of industrial countries and diminish their dependencies on resource rich countries. Other potential sources for the recovery of rare and valuable materials are enriched municipal and industrial solid waste, landfill sites, tailings, and aqueous waste waters (Dodson et al., 2012).

Although during the last years many efforts have been made in numerous countries aiming at e.g. the substitution of rare elements, reducing the environmental impact of mining activities, increasing efficiency of processing technologies, product design taking recycling requirements into account, and progresses in recycling technologies, resulting e.g. in increased recycling rates of e-waste within Europe (Hagelüken et al., 2016) current recycling rates are still rather low for most metals (Zhang et al., 2017). It was estimated that around 15.5 % of the global amount of electronic waste that was produced in 2014 was formally treated (Zhang et al., 2017). High recycling rates exist only for metals mainly used for ferrous and base metals, such as iron and nickel in carbon and stainless steels or copper and aluminum, and for precious metals (mostly from jewelry) (UNEP, 2013). Recycling rates are negligible for more than 30 metals in the periodic table (UNEP, 2011; Zimmermann, 2017), among these are the rare-earth elements (REEs), the platinum group metals (PGMs), indium, gallium, and germanium, that are used in all kinds of high-tech products. In case of REE, that is a group of 17 chemical similar metallic elements that play an essential role in high-tech materials and renewable energies (e.g. permanent magnets in wind turbine generators, rechargeable NiMH batteries, catalysts), recycling rates were less than 1 % in 2011 (Binnemans et al., 2013a; UNEP, 2011). Similarly, the recycling rate of indium, an element that is used in LCD screens, is with around 10 % in 2015 in case of China rather low, although LCD screens are an important secondary source of indium (Wang et al., 2015; Wang, 2009; Zhang et al., 2017). Gallium that is mostly applied in form of GaAs and GaN and used for electronic devices such as integrated circuits or opto-electronic components (e.g. LEDs) is currently not recycled due to numerous recycling barriers (Ueberschaar et al., 2017). Although technical recyclability of PGMs is considered to be very high (> 95 %), recycling rates in Europe are only 5-10 % in electronic devices (e.g. PCs, TVs, mobile phones). However, it has to be mentioned that the main reasons for low recycling rates of e-waste are insufficient collection systems and recycling chains (Bakhiyi et al., 2018; Hagelüken, 2012). Nonetheless, increasing complexity of technical products, inappropriate product design, as well as dissipative usage of metals make an economic technical recyclability more difficult for all elements (Ciacci et al., 2015). Despite the numerous research activities of the last years aiming on the development of effective recycling routes, it is highly probable that recycling rates of these elements are still low. For REE Binnemans et al. (Binnemans et al., 2013b) estimated a recycling rate of 19-35 % in 2020, contributing to 1.3-3 % of the overall supply. Elements that are applied only in minor mass fractions are further diluted in general recycling processes and subsequently get lost. Due to this, many elements such as gallium, indium, thallium, or antimony are currently classified as not recyclable (Ciacci et al., 2015). Therefore innovative approaches that enable a circular economy of metals towards a green economy are still highly demanded.

Biological approaches can contribute to solve some of these problems by enhancing metal recovery from technical waste products, processing wastes, industrial waste waters and other secondary sources. Microorganisms developed many processes that influence biogeochemical cycles of elements, e.g. bio-weathering, microbial reduction, biomineralization and –precipitation, or bioaccumulation. These processes provided by nature can be used as tools for many different clean industrial processes, recovery of metals, novel bio-based materials, bioremediation, but also „green“ recycling processes.

The article gives an overview about modern bio-based approaches that are currently developed for the recycling of value elements from technical products contributing to a „green“ circular economy. Further it presents some new ideas and trends combining the bioeconomy with classical metal processing technologies.

2. Bioleaching: „old“process for new applications

Bioleaching is defined as the extraction of metals by the metabolic activity of bacteria (direct bioleaching) or metabolic compounds (indirect bioleaching). It is applicable to metal extraction from low-grade ores, beneficiation of ores or coal, removal of toxic metals, and recovery of metals from waste materials. Bioleaching has been intensively studied for sulfidic ores using chemolithoautotrophic bacteria e.g. *Acidithiobacillus* spp., *Ferrimicrobium* spp., or *Leptospirillum* spp. (Hutchins et al., 1986; Rawlings, 2002; Vera et al., 2013). These bacteria use ferrous iron and/or reduced sulfur sources as electron donor while producing sulfuric acid and ferric iron as byproducts that contribute to mineral dissolution. The organisms are acidophilic and mostly grow within the pH range 1.5-2.0. These processes are used on a commercial base either in irrigation-type processes (e.g. heap leaching) or in stirred tank-type processes (Rawlings et al., 2003).

In comparison to the numerous studies describing the bioleaching of sulfidic ores, relatively few studies investigate the bioleaching of non-sulphidic ores such as oxides, carbonates, phosphates, and silicates (reviewed by (Jain and Sharma, 2004)). These ores provide no energy source for the microorganisms to utilize. Most of these studies use heterotrophic bacteria and fungi that require an organic carbon source for their growth. These microorganisms produce and secrete metabolites during their fermentation such as organic acids (e.g. citric acid, acetic acid, oxalic acid, gluconic acid, glutamic acid), exopolysaccharides, and siderophores that contribute to dissolution of minerals and complexation of released metal ions. Especially organic acids play a major role in these processes by supplying both protons and a metal complexing organic acid anion (Gadd, 1999). Other molecules are low-molecular or high-molecular weight organic ligands (e.g. oxalic acid or extracellular polysaccharides, respectively) (Gadd, 1999; Welch et al., 1999; Welch and Ullman, 1999) or iron-chelating compounds (siderophores) that increase the rates of mineral dissolution by forming stable soluble metal-organic complexes in solution (Bi et al., 2010; Cheah and Sposito, 1999; Duckworth and Sposito, 2005; Frazier et al., 2004; Hersman et al., 1995; Liermann et al., 2000; Yoshida et al., 2002).

Several factors affect the efficiency of heterotrophic bioleaching processes that have to be considered for process development. Metal release is influenced by applied biomass concentration, metal tolerance of microorganisms, type and amount of metabolic products released into the medium, physicochemical parameters such as temperature, pH, oxygen supply, stirring rate, contact time, composition of media, and properties of material to be leached such as pulp density, grain size, mineral composition, surface properties, and pretreatment (e.g. heating) and has to be investigated for each application (Bosecker, 1997; Guezennec et al., 2017; Olson et al., 2003; Rohwerder et al., 2003; Spolaore et al., 2011).

Another bioleaching approach is biocyanidation that uses biogenic cyanide for recovery of gold or other noble metals. Principally cyanide has been utilized as lixiviant for gold in the mining industries for more than one century (Hilson and Monhemius, 2006). Cyanide is dissolving gold and other noble metals vial an electrochemical process forming metal cyanide complexes. The reaction takes place at pH 10-10.5. Generally, these processes can be applied also for recovery of noble metals from electronic wastes (Cui and Zhang, 2008). However, cyanide is highly toxic. Several accidents in mining industries followed by severe environmental damages caused much concern over the use of cyanide as leach reagent (Hilson and Monhemius, 2006). The use of biogenic cyanide is considered to be a more environmentally friendly approach because extremely lower concentrations than conventional processes are produced and applied (Campbell et al., 2001). Some bacteria e.g. the facultative anaerobic *Chromobacterium violaceum* as the most prominent representative generating cyanide when growing on an appropriate substrate such as glycine at the early stationary phase (Knowles, 1976; Michaels and Corpe, 1965; Michaels et al., 1965). Biogenic cyanide has been discussed to be applied for gold recovery in mining industry (e.g. in heaps, in continuous feed bioreactors at remote mining sites) or gold recovery from electronic wastes (Campbell et al., 2001; Chi et al., 2011; Faramarzi et al., 2004).

Microbial reductive processes are other alternatives that mediate mineral dissolution and metal release. The underlying mechanisms are based on either indirect reduction (production of reducing agents) or direct reduction (enzymatically catalyzed requiring cell contact) (Eisele and Gabby, 2014). For example, iron solubility can be increased by the reduction of Fe(III) to Fe(II) e.g. by extracellular organic ligands that are strong reducing agents (e.g. oxalic acid) while keeping their structure (Taxiarchou et al., 1997). Saal et al (2010). used a mixture of small organic acids (citrate, oxalate) and the siderophore desferrioxamine B for the dissolution of manganese oxide (Saal and Duckworth, 2010). It was supposed that these mixtures enhance both reductive and complex-forming dissolution rates. Other organic compounds e.g. humic acids can act as electron shuttles. These compounds can be reduced by bacteria (e.g. *Geobacter metallireducens* and *Shewanella algae*). The reduced form itself reduces Fe(III) to Fe(II) (Lovley et al., 1996). Some microorganisms, e.g. strains of *S. putrefaciens*, *S. oneidensis*, or *G. metallireducens* obtain energy for anaerobic growth by using Mn(IV) or Fe(III) as their electron acceptors (Dichristina, 1992; Lovley et al., 1993; Myers and Nealson, 1988). These microorganisms oxidize organic carbon (e.g. acetate) to carbon dioxide coupled with the reduction of Fe(III) or Mn(IV) oxide(Lovley et al., 1993; Nealson and Myers, 1992). Several strains of *Thermoanaerobacter* were isolated from deep subsurface environments that use several organic carbon sources or hydrogen as electron donors while reducing various elements such as Fe(III), Co(III), Cr(VI), Mn(IV), and U(VI) at 60 °C followed by secondary mineral precipitation (Roh et al., 2002). In case of all strains it was proposed that the reduction is an enzymatic process. Ehrlich et al. described Mn(IV)-reducing bacteria on manganese nodules that reduce insoluble Mn(IV) to soluble Mn(II) at aerobic or anaerobic conditions (Ehrlich, 2000). These nodules are potentially important natural sources of critical high-tech metals such as Co, Ni, V, W, Nb, Pt, and rare earth elements including yttrium (Guan et al., 2017; Hein et al., 2015). The reduction of Mn(IV) oxide from manganese nodules resulted also in the release of other elements such as Cu, Ni, and Co. Consequently, microbial reduction was suggested to be exploitable for bioleaching of marine nodules (Ehrlich, 2000). Lee et al. used a mixed culture enriched with Fe-reducing microorganisms for bioleaching of Mn, Co, and Ni and obtained a leaching efficiency of > 70 % for all elements (Lee et al., 2001). In more recent publications the bacteria *Shewanella putrefaciens* and *Aeromonas hydrophila* were used for bioleaching of jarosites and gossan minerals (Castro et al., 2016, 2017). Bioleaching activity was based on the reduction of Fe(III) that is insoluble at neutral pH. The strain *S. putrefaciens* CN2 used some of the added jarosites as electron acceptor leading to a relatively high solubilization of Fe(III) of > 80 %. In another experiment the mineral dissolution was stimulated by the addition of Fe(III) chelators (e.g. citrate) leading to a reduction yield of > 90% of Fe(III) (Castro et al., 2017). Similarly, *A. hydrophila* used different biogenic jarosites as electron acceptor followed by a release of Fe(II). The cells attached through pili to the mineral surfaces, which were used for the electron transfer (Castro et al., 2016). The solubilization processes were accompanied by secondary mineralization leading to the formation of siderite, vivianite, or magnetite.Bioleaching approaches were firstly developed for metal extraction from ores and current commercial usage concentrate on these applications. However, the processes can be principally applied also to other anthropogenic sources such as mining residues, industrial residues and technical waste materials that can be considered as an artificial ore. During the last years a growing number of studies were published that describe the application of bioleaching approaches for the recovery of metals from various technical and industrial waste products, e.g. fly ash from municipal waste incineration, electronic scrap such as printed circuit boards from computers and mobile phones, spent catalysts and batteries and others partly reviewed by Lee (Lee and Pandey, 2012). Some examples of these studies are summarized in Table 1, including some more recent studies, while covering a broad spectrum of elements (including REE), materials, and approaches. These examples demonstrate a high diversity of research strategies as well as the high innovation potential in this field. The studies are based on the use of chemolithoautotrophic bacteria, heterotrophic bacteria, yeasts and fungi, and cyanogenic bacteria. Most studies with chemolithoautotrophic bacteria used the acidophilic *A. ferrooxidans* or *A. thiooxidans*. These approaches obtained relatively high leaching efficiencies (in many cases mobilization of > 80 % of metals) but required the addition of sulfur or Fe(II) and acidification of the cultivation media in order to maintain bacterial growth and solubilization process (Bayat et al., 2009; Brandl et al., 2001; Brombacher et al., 1998; Cerruti et al., 1998; Funari et al., 2017; Gholami et al., 2011; Karwowska et al., 2014; Klink et al., 2016; Mishra et al., 2008; Oliazadeh et al., 2006; Willscher et al., 2007; Zhao et al., 2008). Studies describing approaches with heterotrophic microorganisms used bacteria, fungi or yeasts that produces diverse organic acids such as citric acid, gluconic acid, or acetic acid as lixiviant for the respective metals. Especially the fungus *Aspergillus niger* was used for a broad range of materials (Bosshard et al., 1996; Brandl et al., 2001; Jadhav et al., 2016; Kim et al., 2016; Qu et al., 2015). Depending on growth conditions, this organism produces huge amounts of diverse organic acids (e.g. citric acid, gluconic acid, oxalic acid). The organism is commercially applied for organic acid production and its cultivation as well as its metabolism have been studied in detail. In some cases more than 90 % of metals could be mobilized by application of this organism . Biogenic cyanide produced by *Chromobacter violaceum* was used for the mobilization of gold and other noble metals, copper, and nickel from shredded printed circuit boards and automotive catalysts by using direct or indirect leaching approaches (Campbell et al., 2001; Chi et al., 2011; Faramarzi et al., 2004; Shin et al., 2015). In indirect bioleaching processes more than 90 % of metals were mobilized. These results were comparable with the usage of commercial NaCN demonstrating the principle suitability of such approaches for commercial applications (Shin et al., 2015). Remarkably, recent developments show that more complex processes that include consortia of different bacteria, isolation and application of new microbial strains, pretreatment of waste materials, combination with chemical methods or ingredients, or sophisticated bioprocesses become increasingly important (Funari et al., 2017; Jadhav et al., 2016; Kim et al., 2016; Pant et al., 2014; Wang et al., 2016; Zhao et al., 2008). Such new approaches bear the potential for higher leaching rates and extended substrate spectra that are important for an commercial utilization.

Most studies aimed at the recovery of value metals such as Cu, Ni, Au, or other noble metals that can be found in high concentrations in diverse electronic wastes or in residues from mineral processing e.g. smelter dust. However, starting with the resource crisis in 2009, some studies were published that concentrate on other value elements, e.g. rare earth elements (Hopfe et al., 2017; Qu et al., 2015; Qu and Lian, 2013; Reed et al., 2016). It can be expected that the ongoing growing demand of other elements, development and growth of new technologies such as renewable energies will promote further studies.

One major issue that influences the bioleachability of metals is the toxicity of elements. This differs depending on the organism. The toxicity had a direct influence on the optimal pulp density, limit the applied pulp density or favor indirect leaching over direct leaching processes. The growing use of indigenous bacteria adapted to the environments instead of well-known strains from lab collections may overcome these limitations.

Table 1: Bioleaching of industrial waste and scrap materials

Material	Organism	Leached metal	Results and remarks	Mechanism	Reference
Electronic waste; Printed Circuit Boards	*Acidithiobacillus ferrooxidans, A. thiooxidans*	Cu, Sn, Al, Ni, Pb, Zn	Dust from electronic waste recycling; Mobilization of > 90 % of Cu, Al, Ni, Zn; addition of elemental sulfur required	Sulfuric acid and ferric iron	(Brandl et al., 2001)
	Penicillium chrysogenum	Cu, Al, Zn, Ni, Fe, Pb	> 90 % of Cu depending on pulp density and leaching mode	Organic acids (citric and gluconic acid)	(Ilyas and Lee, 2013)
	Consortium of different bacteria with *Alicyclobacillus* spp. *and Sufobacillus* spp. as dominant species	Cu	> 95 % of Cu	Fe-mediated bioleaching; addition of biochar	(Wang et al., 2016)
	Aspergillus niger	19 elements	100 % solubilization of all metals	Pretreatment with NaOH; combination of organic acids and hydrogen peroxide	(Jadhav et al., 2016)
	Chromobacterium violaceum	Au	appr. 15 % leaching efficiency	Biogenic cyanide	(Faramarzi et al., 2004)
	Chromobacterium violaceum	Au, Cu	24.6 % and 11.3 % leaching efficiency for Cu and Au, respectively	Biogenic cyanide	(Chi et al., 2011)
	Consortium of sulfur oxidizing bacteria (e.g. *Acidithiobacillus ferrooxidans*), partly with surfactin producing bacteria (e.g. *Bacillus subtilis*)	Cu, Cd, Cr, Ni, Pb, Zn	48.5 % of Cu with acidic approaches; addition of sulfur required	Sulfuric acid	(Karwowska et al., 2014)
	Consortium with *Leptospirillum ferriphilum* and *Acidithiobacillus caldus* as dominant species	Cu, Al, Zn, Pb, Sn	> 75 % of Cu; addition of sulfur	Sulfuric acid and ferric iron	(Xia et al., 2017)
	Consortium of acidophilic iron and sulfur oxidizers and heterotrophic bacteria	Cu, Zn, Cr, Ni, Pb, Mn	> 90 % of Cu, > 40 % of Zn; formation of secondary minerals	Sulfuric acid and ferric iron	(Willscher et al., 2007)
Fly ash from municipal waste incineration	*Aspergillus niger*	Cd, Zn, Cu, Pb, and others	Up to 81 % Cd, 66 % Zn, 57 % Cu, 52 % Pb recovery; pulp density of 7 %	Organic acids (gluconic acid)	(Bosshard et al., 1996)

	Acidithiobacillus ferrooxidans	Cd, Cu, Mn, Ni, Zn, and others	> 80 % Cd, Cu, and Zn recovery; pulp density of 5 %	Sulfuric acid and ferric iron	(Brombacher et al., 1998)
	Mixed acidophilic culture comprising *Aciditiobacillus ferrooxidans, thiooxidans, caldus* and others	24 elements e.g. Cu, Ce, Nd, Ga	Comparison with chemical leaching; leaching yields > 85 % for a number of elements; bioleaching with lower H_2SO_4 consumption	Sulfuric acid	(Funari et al., 2017)
	Indigenous *Alkalibacterium* strain and other alkaliphilic strains	Cu	Up to 52 % Cu recovery; tolerates high pulp densities up to 20 %	metabolites	(Ramanathan and Ting, 2016)
Spent automotive catalyst	*Chromobacterium violaceum*	Pt, Pd, and Rh	> 90 % leaching efficiency by indirect bioleaching, comparable to leaching with NaCN	Biogenic cyanide	(Shin et al., 2015)
Fluorescent lamp phosphor from spent lamps	Kombucha „tea fungus“ (consortium of *Zygosaccharomyces lentus* and *Komagataeibacter hansenii)*	Rare Earth Elements (Y, Eu)	Up to 7.4 % of total REE, 12.5 % of Y	Organic acids	(Hopfe et al., 2017)
	Gluconobacter oxydans	Rare Earth Elements	About 2 % of total REE	Gluconic acid and other organic acids	(Reed et al., 2016)
Cracking catalysts	*Gluconobacter oxydans*	Rare Earth Elements	About 49 % of the total REE	Gluconic acid and other organic acids	(Reed et al., 2016)
Red mud	*Penicillium tricolor, Aspergillus niger*	Rare Earth Elements, Ga, Ge, V	> 40 % leaching efficiency for e.g. Ga, Ge, Yb	Organic acids (oxalic and citric acid)	(Qu et al., 2015; Qu and Lian, 2013)
Cathode ray tube waste	*Serratia plymuthica*	Pb, Ba, Ca, Si, Cd	Release of all elements at alkaline conditions; addition of EDTA	Complexation (e.g. siderophores), formation of hydrous oxides	(Pant et al., 2014)
Spent lithium ion batteries	*Aspergillus niger*	Cu, Li, Mn, Al, Co, Ni	> 90 % leaching efficiency for Cu and Li, depending on approach	Organic acids (citric acid, oxalic acid)	(Horeh et al., 2016)
	Acidithiobacillus ferrooxidans	Co, Li	Appr. 65 % leaching efficiency for Co and 10 % for Li	Sulfuric acid and ferric iron (produced from elemental sulfur and ferrous iron)	(Mishra et al., 2008)
spent Mo-Co-Ni refinery catalyst	*Acidithiobacillus ferrooxidans*	Co, Mo, Ni	recovery of 63 % Al, 96 % Co, 84 % Mo, and 99 % Ni up to 75 % of Ni	Sulfuric acid and ferric iron (produced from ferrous sulphate)	(Gholami et al., 2011)

Copper smelters flue dust	*Acidianus ferrooxidans* and *Acidianus thiooxidans*	Cu	87 % leaching efficiency for Cu	Sulfur oxidation	(Oliazadeh et al., 2006)
	Acidithiobacillus ferrooxidans and *Acidiphilium* spec.	Re, Sb, Co, Ge, Zn, Cu	45 % and 70 % leaching efficiency for Cu and Zn	Sulfuric acid and ferric iron	(Klink et al., 2016)
Spent Ni-Cd batteries	*Acidithiobacillus ferrooxidans*	Cd, Ni, Fe	Bioreactor for bacterial cultivation and bioleaching reactor were seperated; recovery of 100 % Cd, > 90 % of Ni and Fe	Sulfuric acid (elemental sulfur as energy source)	(Cerruti et al., 1998)
	Consortium of *Acidithiobacillus* strains	Ni, Cd	Continuous flow two-step system; up to 100 % leaching efficiency for Ni and Cd	Sulfuric acid	(Zhao et al., 2008)
	Aspergillus niger	Ni, Cd, Co, Zn	> 90 % Ni and Cd recovery	Organic acids (oxalic and citric acid)	(Kim et al., 2016)
Steel plant waste	*Acidithiobacillus ferrooxidans*	Zn, Fe	35 % and 37 % leaching efficiency for Zn and Fe, respectively	Sulfuric acid	(Bayat et al., 2009)

However, the dissolution of metals is only one first step in metal recovery. A major challenge is the cost-effective and selective recovery of low concentrations of metals from large volumes of dilute leachates. Principally, different technologies can be applied to achieve this goal. Most of these are classical approaches such as cementation, solvent extraction, ion exchange, chemical precipitation, or adsorption on activated carbon (reviewed by (Cui and Zhang, 2008)). Newer approaches include sophisticated membrane technologies and dialysis (e.g. (Zeidler, 2014)), novel materials for adsorption, electrowinning (reviewed by (Moats and Free, 2007)), but also biobased approaches such as biosorption (chapter 3), bioreduction (chapter 5), microbial electrochemistry (reviewed by (Modin and Aulenta, 2017; Nancharaiah et al., 2016)) or bioprecipitation (e.g. (Fang et al., 2011)). For example, most recently Roosen et al. (Roosen et al., 2017) developed a new adsorbent material based on alginate-sulfonate-silica particles and used it for the removal of indium from leachates. Park et al. (Park et al., 2017) used *E. coli* cells expressing lanthanide binding tags on their surface for the removal of Rare Earth Elements from leachates. However, these approaches are in an early stage of research. Modin et al. (Modin et al., 2012) described a bioelectrochemical method for the selective recovery of Cu, Pb, Cd, and Zn from diluted solutions that could potentially applied for leachates.

Despite these progresses, there are still some obstacles that have to be considered, such as low selectivity of metal recovery especially in case of complex solutions, use of toxic chemicals, high economic costs, and generally difficulties to recover elements from highly diluted solutions. However, it can be expected that future trends overcome some of these limitations though it will still be some time before these technologies are commercialized.

3. Biosorption: selective recovery of elements from industrial waters

Biosorption is defined as the property of biomass or certain biomolecules to bind and concentrate selected ions or other molecules from aqueous solutions (Volesky, 2007). It is a passive process and independent from metabolic activities. Therefore nutrients are not required and processes can performed in environments with high toxicity. Biosorption has been mainly applied for the removal of toxic metals from polluted waters, such as arsenic, chromate, cadmium, or uranium (Volesky and Holan, 1995).

Another attractive application is the recovery of value metals such as gold, platinum, palladium or others from solutions (Das, 2010; Pollmann et al., 2006b). Conventional pyrometallurgical or hydrometallurgical methods (e.g. adsorption by ion exchange resin, activated carbon, or minerals, solvent extraction, chemical precipitation) require high amounts of energy, addition of chemical agents thus generating secondary wastes, or are inefficient especially for highly diluted solutions (Das, 2010). Biosorption is an environmentally friendly alternative to these methods because it uses biodegradable compounds that can be easily produced in high amounts. Further, biomass is considered as carbon-neutral and petrochemical independent as it does not emit extra carbon dioxide when burned (Maruyama et al., 2007; Ritter, 2004). Various types of biomass have been reported to bind and concentrate metal ions from industrial effluents and aqueous solutions (Dodson et al., 2015; He and Chen, 2014; Nancharaiah et al., 2016).

Metal containing solutions such as industrial waste waters, leachates, and mining waters are often acidic with pH < 3, have a complex composition containing different competing elements, and contain toxic chemicals or organic compounds that influence biosorptive properties. Therefore, major challenges of biosorptive approaches are stability of materials, selectivity, effectivity, and cost efficiency. There are several approaches that address these challenges. Most studies concentrated on the use of bacterial cells, fungi, yeast (Volesky and Holan, 1995), algae (reviewed by (He and Chen, 2014)), seaweed biomass (Figueira et al., 2000) or biocomponents such as crab shells (Daubert and Brennan, 2007), plant fibers (Salamun et al., 2015), chicken feathers (Ishikawa et al., 2004) etc. as biosorptive components that can be easily produced or are waste materials (e.g. in case of crab shells) (reviewed by (Dodson et al., 2015)). Especially biopolymers such as cellulose, chitin, or chitosan materials are chemically resistant. However, these materials possess no selectivity and bind a broad range of different elements. This is a drawback for their application in metal recovery processes because these applications intend not only the concentration of metals from highly diluted solutions but also the selective recovery of metals of interest.

Therefore more recent developments concentrate on the use of defined proteins from biomass or the direct engineering of improved microbes and enzymes. Marayuma et al. (2007) tested different model peptides, proteins, and protein-rich biomass regarding their capability to selectively bind different precious metals from model solutions, metal refining solutions, and industrial wastes at acidic conditions (Maruyama et al., 2007). All tested biomass as well as proteins selectively adsorb Pd and Au ions in the presence of transition elements. Further, it was possible to remove Au, Pd, and Pt from plating wastes using protein-rich chicken egg-shell membrane.

Different approaches use the metal binding motifs of natural proteins, e.g. metallothioneins, as biosorptive component. Metallothioneins (MT) are cysteine-rich proteins that bind different metals such as Cd, Hg, Cu and Pb. MTs from different natural sources have been expressed in *E. coli* and *Pseudomonas putida* and used as biosorbent, mainly for removal of heavy metals (reviewed by (Chen et al., 1999) and (Mejare and Bulow, 2001)). However, these proteins are also attractive for the recovery of value metals. Terashima et al. (2002) produced a fusion protein composed of the maltose binding protein and human MT and immobilized it on Chitopearl resins (Terashima et al., 2002). These materials were used for binding of Cd and Ga in a concentration range of 0.2–1.0 mM. Further, the biosorbents could be used several times without loss of binding activity.

Recently, CadR that is a Cd-binding protein first isolated from *Pseudomonas putida* has been expressed on the surface of *E. coli* cells. These engineered cells show a high Cd^{2+} adsorption capacity of 19.5 µmol Cd(II) g^{-1} cells (Liu et al., 2015).

Proteinaceous bacterial surface layers that envelope many bacterial cells are other interesting biomolecules that have been used for the binding of different elements such as U, Pd, Au, or Cu (Allievi et al., 2011; Merroun et al., 2005; Pollmann et al., 2006b). The binding of U, Pd, and Au has been investigated in more detail in case of the S-layers from *Lysinibacillus sphaericus* JG-A12 and NCTC 9602 (Fahmy et al., 2006; Jankowski et al., 2010; Merroun et al., 2005). These elements were coordinated by phosphate and carboxyl groups (Fahmy et al., 2006; Merroun et al., 2005), in case of Au(III) it was assumed that amine groups were involved in complexation (Jankowski et al., 2010). Due to their self-assembling properties that enable the formation of nanostructured protein arrays on various technical surfaces (Sleytr et al., 2014; Toca-Herrera et al., 2005; Weinert et al., 2015b), S-layer proteins are attractive biomolecules for the construction of biosorptive composites (Suhr et al., 2014). For example, so-called biocers were produced by entrapping S-layer carrying cells or S-layers in porous ceramics using sol-gel technology and used for the removal of U from contaminated waters (Raff et al., 2003; Soltmann et al., 2002). Pollmann et al. (2007) constructed modified His-tagged S-layer proteins that exhibited enhanced Ni-binding capacities while self-assembling to a nanoporous protein meshwork (Pollmann and Matys, 2007).

Peptides are other, less complex and easily synthesizable biomolecules that have been used for the design of various biosorbents. Stair et al. (2005) synthesized and immobilized various peptides of different lengths composed of Gly, Asp, and Cys residues on commercial Tentagel resins and used it as biosorbents for the binding of Ni^{2+}, Cd^{2+}, Co^{2+}, and Mg^{2+} (Stair and Holcombe, 2005).

A novel very promising approach is the selection of metal binding peptides by phage surface display. With this technique, peptides selective for several metallic surfaces or metal ions were identified (Sarikaya et al., 2003; Seker and Demir, 2011). Currently these peptides are mainly used for the development of nano-materials and composites, but they are also attractive as biosorbents. Nian et al. (2010) selected Pb^{2+}-specific peptides via a chromatographic biopanning methods from phage display libraries (Nian et al., 2010). They identified one bacteriophage expressed peptide (TNTLSNN) with high affinity and specifity to Pb^{2+} as proved by cross-binding assay to different metal ions. In a follow-up study Nguyen et al. (2013) constructed a recombinant *E. coli* displaying the peptide on its cell surface thus obtaining a highly selective *E. coli*-based biosorbent (Nguyen et al., 2013). Similarly, Yang et al. (2015) selected Cr(III) binding phages from a phage display library (Yang et al., 2015). A phage expressing the heptameric peptide YKASLIT was immobilized on cytopore beads for Cr(III) preconcentration. Most recently, Sawada et al. (2016) selected Nd(III) binding bacteriophages via phage surface display technology (Sawada et al., 2016). These phages were used as adsorbent for the selective recovery of Nd(III) from mixed solutions of Nd(III) and Fe(III), mimicking the dissolved solution of neodymium-iron-boron alloys ($Nd_2Fe_{15}B$) indicating a high potential to be applied in recycling strategies. In another approach, lanthanide oxide particles were used as target to select peptides that induces the precipitation of lanthanide hydroxides (Hatanaka et al., 2017). Three peptides (SCLWGDVSELDFLCS, SCLYPSWSDYAFCS, SCPVWFSDVGDFMVCS) were identified that mediate the mineralization of lanthanide ions. The researchers proposed that such peptides have a potential for the separation of lanthanides via selective mineralization.

Yunus et al (2015) immobilized genetically engineered fusion proteins composed of palladium binding peptides and cellulose binding domains on cellulose materials (Yunus and Tsai, 2015). These constructs were used as biosorbents for the selective binding of Pd(II). The materials were able to selectively bind Pd(II) from a mixture of Pd(II) and Pt(IV) with a maximum adsorption capacity of 175.44 mg/g. Further, it was possible to remove the bound Pd and reuse the biosorbent several times without losing the binding capacity. The materials were working at a wide range of different pH (pH 1.8-11) and temperatures (10-40 °C), therefore they can be applied at different conditions. These examples demonstrate that phage surface display technology is a promising strategy to identify highly selective peptides for different elements that can be used for the construction of biosorbents not only for bioremediation but also for the recovery of value metals.

In numerous studies metal binding motifs and peptides were expressed and anchored on the surface of microbial cells via fusion with outer membrane proteins. In many approaches the peptides were anchored to the outer membrane protein LamB, thus obtaining engineered microbes that worked as efficient adsorbent. For example, hexa-His chains were expressed on the surface of *E. coli* by construction LamB hybrids. These cells exhibited high affinity to Ni ions (Sousa et al., 1996). Other researchers expressed metallothioneins or metal binding peptides as fusions to membrane or membrane associated proteins in *E. coli*, *P. putida*, yeasts, or other microorganisms (Kotrba et al., 1999a; Kotrba et al., 1999b; Nishitani et al., 2010; Sousa et al., 1998; Valls et al., 2000a; Valls et al., 2000b; Valls et al., 1998). In a very recent publication Park et al. (2016) produced fusion proteins comprising the surface (S-layer) protein of *Caulobacter crescentus* and peptides that have been used as lanthanide binding tags for protein purification, biosensing, and NMR spectroscopy (Liang et al., 2013; Martin et al., 2007; Nitz et al., 2003; Park et al., 2016). These hybrid proteins were expressed in the cell surface of *C. crescentus* in high density (Park et al., 2016). The engineered cells exhibited an enhanced sorption of REE and a high specifity for REE. Further, it was possible to desorb the bound REE enabling a repeatable reuse of the bioadsorbents. The same tags were used for surface expression on *E. coli* cells using the *omp*A system (Park et al., 2017). These constructs were successfully used to remove REE from highly diluted low-grade feedstock leachates. Thus, cell surface display is an attractive approach for implementation in recycling processes.

Other approaches use metabolic products as complexing agents. Very interesting biomolecules are siderophores. These small organic molecules are iron chelators that are produced and secreted by bacteria and are used for the uptake of iron. In addition to iron, other metals, e.g. Ga, Co, different actinides, can be complexed by the siderophores (Brainard et al., 1992; Gascoyne et al., 1991a, b; Harrington et al., 2012). These properties make them attractive for biotechnological applications. Biopolymers are another attractive group of metabolites. Bacterial poly(γ-glutamic acid) has been used for the adsorption of toxic Hg(II) (Inbaraj et al., 2009), Pb(II) (Mu et al., 2011), and Fe(III) (Bodnar et al., 2013). Varshini (2015) constructed a modified biohydrogel and used it for the removal of the Rare Earth Element Ce(III) from industrial effluents (Varshini et al., 2015). Different extracellular polymeric substances have been used for the removal of Co(II), Cu(II), and other elements (Dobrowolski et al., 2017; Mona and Kaushik, 2015; Perez et al., 2008). Natural polysaccharides such as alginate, chitin, chitosan, starch as well as derivatives of them and polysaccharide based composites have been widely used for the removal of different heavy metals (reviewed by (Crini, 2005)).

Most studies on biosorption of value elements consider the recovery of precious metals, e.g. Au, Pd, and Pt from solutions. Only few recent studies investigate the biosorption of other value elements, e.g. of Rare Earth Elements (REE) although the molecular interactions of these elements with certain kind of biomolecules have been studied. Especially phosphoproteins and Ca-binding proteins are well known for their interaction with REE.

These properties have been used for the enrichment of these proteins (Chaga et al., 1996; Guzel et al., 2012). Minoda et al. (2015) used the red alga *Galdieria sulfuraria* for the recovery of REE from a mixture of different elements (Minoda et al., 2015). Other researchers studied the adsorption of lanthanides by diverse microorganisms (e.g. *Bacillus subtilis* (Markai et al., 2003; Moriwaki et al., 2013), *P. fluorescens* (Suzuki et al., 2005), *Streptomyces acidiscabis* (Haferburg et al., 2007)) (reviewed by (Andres et al., 2003; Moriwaki and Yamamoto, 2013)). In a recent publication Park et al. (2016) expressed lanthanide binding tags, that were originally developed for spectroscopic applications by Nitz et al. (2003) (Franz et al., 2003; Nitz et al., 2003), as S-layer fusion proteins on the surface of *Caulobacter crescentus* (Park et al., 2016). These constructs were used for the adsorption of different REE (e.g. Nd(III), Ce(III), La(III)) from acid leachates. Different microorganisms have been described to accumulate Ga and were used for Ga removal (Gascoyne et al., 1991a). The accumulation is mediated by siderophores that are complexing with Ga. Phosphoproteins and phosphopeptides are other biomolecules that interact with Ga(III). These properties have been used in IMAC procedures for the enrichment of phosphoproteins (Aryal et al., 2008).

In order to enable a low cost usage, the biocompounds should be recycled and reused after adsorption. The repeatable use of biomolecules for biosorption requires the immobilization of the molecules to an appropriate surface. The combination of bio-compounds with inorganic materials brings together advantages of both materials. Soltmann et al. (2003) developed uranium binding composites, so called bio-ceramics (biocers), by immobilization of bacterial cells or surface (S) layer proteins via sol-gel techniques (Soltmann et al., 2002). These composites were used for the removal of uranium from waters but also for the binding of Pd(II) and copper (Pollmann et al., 2006a; Pollmann et al., 2006b; Raff et al., 2003). Yunus et al. (2015) immobilized fusion proteins composed of palladium binding peptides and cellulose binding domains on cellulose (Yunus and Tsai, 2015). These complexes were used for the adsorption of Pd(II) from model solutions at various conditions. In addition, it was possible to desorb the Pd(II) using 1 M thiourea thus creating a reusable Pd(II) selective biosorbent. Other approaches entrap the biocompounds in polyvinyl alcohol, chitosan, hydrogels, or alginate (Ting and Sun, 2000).

4. Bioflotation: separation of particles with biological means

Microbial cells, cell components, metabolites or other biomolecules can interact with solid substrates and modify surface properties, e.g. by introducing hydrophobic properties by adhesion to the surfaces (Das et al., 1999; Patra and Natarajan, 2006). These properties can be used for mineral beneficiation. For example such biocompounds have been reported as environmentally friendly collectors or depressants and were applied as flotation reagents in selective mineral separation (reviewed by (Behera and Mulaba-Bafubiandi, 2017)). Most of these approaches concentrate on the use of bacterial cells or their products that have been described to specifically interact with minerals. For example, acidophilic sulphide oxidizing bacteria such as *Acidithiobacillus ferrooxidans* that have been widely applied for bioleaching of sulphidic minerals selectively attach to sulphide surfaces while forming a biofilm (Harneit et al., 2006). These interactions have been studied on molecular level (Florian et al., 2010; Gehrke et al., 1998; Harneit et al., 2006). The cells have naturally a high affinity for sulphide minerals. Further, it was found that the surface chemistry of *A. ferrooxidans* cells altered in the presence of sulfur, pyrite or chalcopyrite (Das et al., 1999). Consequently, these bacteria have been identified as promising candidates for their application in bioflotation processes. In several studies the cells were used for the depression of pyrite (Das et al., 1999; Hosseini et al., 2005; Mehrabani et al., 2011; Sharma et al., 1999). For example, the application of *A. ferrooxidans* was discussed as replacement of toxic NaCN in the flotation of high-pyrite, low-grade lead-zinc ore (Mehrabani et al., 2011). Other organisms that have been investigated for their use in flotation processes are *Rhodococcus* strains for the flotation of copper oxide minerals (Kim et al., 2017) or as collector for hematite (Olivera et al., 2017; Yang et al., 2013) or the separation of apatite and quartz (Merma et al., 2013), the yeast *Saccharomyces cerevisiae* for the separation of quartz from hematite or calcite by rendering quartz surfaces hydrophobic (Natarajan and Padukone, 2012; Padukone and Natarajan, 2011), *Paenibacillus polymyxa* for the selective separation of pyrite and galena or pyrite and chalcopyrite from quartz and calcite (Patra and Natarajan, 2004, 2006), *Bacillus subtilis* for the separation of pyrite from galena (Sarvamangala et al., 2013), and others (recently reviewed by (Behera and Mulaba-Bafubiandi, 2017)).

Attachment of bacterial cells to surfaces and biofilm formation are mediated by biopolymers and so-called extracellular polymeric substances (EPS) (Gehrke et al., 1998; Kinzler et al., 2003; Vu et al., 2009). They form the structure and architecture of the biofilm matrix. The EPS are composed of an undefined complex mixture of biopolymers primarily consisting of polysaccharides, but also lipids, proteins, humic acids, and nucleic acids (reviewed by (Vu et al., 2009)). Their composition depends on type of microorganisms, age of biofilm, and environmental conditions, including surface properties (Donlan, 2002). Their affinity to surfaces make them interesting for their application in flotation processes. Consequently several studies investigated the effect of EPS as bioreagent in mineral separation. Govender & Gericke (2011) studied the use of EPS extracted from bioleaching consortia for the flotation of a mixture of chalcopyrite and pyrite (Govender and Gericke, 2011). They reached a recovery rate of 70 % of chalcopyrite in comparison to 32 % when using sodium isobutyl xanthate.

Besides EPS other biomolecules has been investigated regarding their application in mineral separation. Especially biosurfactants are interesting compounds that have been applied as frothers in many flotation experiments. Biosurfactants are surface active organic molecules that are produced by many microorganisms. They are lowering the surface tension at the interfaces of solid, liquid, and gases. In contrast to common commercial chemical surfactants they are less toxic, biodegradable, and effective under extreme conditions. Consequently, there are numerous potential fields of application, including pharmaceutical industry, environmental remediation and petroleum industry (recently reviewed by (Saha and Rao, 2017)). Khoshdast et al. (2012) used rhamnolipid biosurfactants produced by *Pseudomonas aeruginosa* as frothing agent in the flotation of copper ores (Khoshdast et al., 2012a; Khoshdast et al., 2012b). It was found that the rhamnolipid was more surface active than the commercial frothers MIBC and DF-250 and the flotation of iron minerals was improved. Another study used biosurfactants produced by *Bacillus circulans* and *Streptomyces* sp. for the flotation of serpentinite and quartz (Didyk and Sadowski, 2012). Vecino et al. (2013) investigated the adsorption of biosurfactants produced by *Lactobacillus pentosus* onto river sediments and suggested their application as a potential foaming agent in froth flotation (Vecino et al., 2013). In both publications the produced biosurfactants were not further characterized.

Vasanthakumar et al. investigated the effect of intact cells, thermolyzed cells, and different fractionated cellular components of *B. circulans* on flotation of sphalerite from a sphalerite-galena mineral mixture (Vasanthakumar et al., 2012). They identified DNA as key constituent responsible for the flotation of sphalerite. It was demonstrated that double stranded extracellular DNA (eDNA) that are secreted by the bacteria on their cell surface as well as genomic double stranded (dsDNA) and single stranded DNA (ssDNA) acted as biocollector whereas other components interacted with galena and worked as depressant. In the presence of ssDNA and other components a maximum flotation recovery of sphalerite of 85 % was achieved. The authors assumed that the amphipathic nature of ssDNA comprising polyphosphate groups on one side and hydrophobic bases on the opposite side facilitated the flotation process. Similar results were obtained for *B. megaterium* (Vasanthakumar et al., 2014).

Fig. 1: Use of extracellular polymeric substances (EPS) or EPS producing bacteria for particle separation. EPS are interacting with fine particles.

All these approaches include living cells or natural biocompounds that were isolated from living cells. Another attractive approach is the use of mineral-specific phages or peptides that were selected from large phage library constructs using biopanning procedures or that were specifically designed. In these studies phages were selected displaying peptides that bind selectively to sphalerite and chalcopyrite (Curtis et al., 2009), chalcopyrite and pyrite (Dunbar et al., 2008), and chalcopyrite and enargite (Curtis et al., 2017) on their surfaces. The interaction of chalcopyrite specific phages with the mineral surfaces was studied in more detail with spectroscopic methods and it was found that the binding of phages altered the surface properties of chalcopyrite. Further, in a mixture with silica the binding of phages induced an aggregation of chalcopyrite particles, proving their principle suitability as agent for mineral separation (Curtis et al., 2011). Currently these procedures are too expensive for an industrial application but are an interesting perspective for new bioreagents that can replace existing flotation agents.

The previous examples were specifically developed for mineral beneficiation and were not applied for recycling purposes. However, principally these developments could be transferred to the separation of fine particles that are released during recycling processes and cannot be targeted by existing processes. Most recently Lederer et al. (2017) (Lederer et al., 2017) described the selection of phages displaying $LaPO_4:Ce^{3+},Tb^{3+}$ (LAP) specific peptides on their surface. Moreover the researchers modified the amino acid sequence and reached a > 5000 fold higher binding strength to LAP in comparison to the wild-type. The directed modification of individual amino acids was proven to increase or decrease the binding specificity and affinity of a peptide to the target material drastically. These phages bound to several components of compact fluorescent lamps (LAP, $CeMgAl_{11}O_{19}:Tb^{3+}$ (CAT)), but not or only weak to $Y_2O_3:Eu^{3+}$ (YOX), $LaPO_4$, SiO_2, and $BaMgAl_{10}O_{17}:Eu^{2+}$ (BAM). This proof of principle shows that the researchers are able to identify perfect fitting biomolecules for target particles by using the improved phage surface display techniques. The authors proposed an application as collectors in bioflotation processes for the separation and recycling of fluorescent phosphor components from electronic scrap. The successful application of phage expressing mineral specific peptides in flotation processes was described earlier (Curtis et al., 2009; Curtis et al., 2011; Dunbar et al., 2008; Liao, 2010). The hydrophobic body of phages improves their interaction with hydrophobic gas bubbles. The directed particle transport via phage peptides in a flotation process is possible. Zeta potential measurements have already proven the changed surface charge of particles of interest after phage interaction (Curtis et al., 2017). However, our studies have shown that the high scale amplification of phage particles is highly affected by mutations making a large scale application of phage in flotation processes impossible. These mutations decrease the number of expressed particle specific peptides at the phage surface (Figure 2) and avoid specific LAP binding. Temperature optimized protocols avoid the fast mutation rate of peptides. However, a phage-independent biomolecule application in flotation systems is recommended.

Figure 2. Amplification of different LAP binding phages and their binding performance. Diagram A shows the increase of the phage number after different amplification rounds of three different specific LAP binders. Amplifications were performed identically. Diagram B shows the binding tests of three individual phage clones and four different phage amplificats per clone to LAP and the numbers were compared to the wild type phage.

Other approaches anchored ZnO, Au, or TiO_2 binding peptides or organic molecules to magnetic particles and separated the respective particles from colloidal mixtures (Essinger-Hileman et al., 2013; Shen et al., 2017; Vreuls et al., 2011) (Table 2). Given the high number of peptides that have been described to selectively interact with various inorganic surfaces and that were mostly applied for material syntheses or sensory applications (reviewed by (Care et al., 2015; Seker and Demir, 2011)) one can assume that these separation technologies can be easily transferred to other materials.

Table 2: Biocompounds with specific particle interactions and potential applications for metal recycling

Biocompound	Target material	Research results, applications	Reference
ZnO binding genetically engineered peptides anchored to magnetic beads (Dynabeads M-270)	ZnO powder	Magnetic separation of Cu_2O and ZnO particles; functionalized magnetic beads were reusable; proof of principle, extension to the sorting of all types of inorganic particles was proposed	(Vreuls et al., 2011)
Gold specific peptides anchored to streptavidin-coated Fe_2O_3 nanoparticles	Au nanoparticles	Magnetic separation of Au nanoparticles from a colloidal mixture of Au and CdS nanoparticles; proposed application: colloidal nanoparticle purification	(Essinger-Hileman et al., 2013)
Fluorescent phosphor LaPO4:Ce^{3+},$Tb3^{+}$ (LAP) binding phages	LAP of compact fluorescent lamps	Identification and genetic engineering of LAP binding peptide sequences; proposed application: separation of REE compounds from technical waste products (e.g. compact fluorescent lamps)	(Lederer et al., 2017)
Bisphosphonate-functionalized Fe_3O_4 nanoparticles; chemical synthesis with anchored dopamine as precursor	TiO_2 nanoparticles	Functionalized magnetic nanomaterials were reusable; capturing of TiO_2 nanoparticles from complex sample matrix; proposed application: preventing environmental pollution, recycling of nanosized TiO_2	(Shen et al., 2017)

5. Bioreduction: creation of nanocatalysts from waste

Precious metals of the platinum group metals such as platinum, palladium, rhodium, ruthenium are widely used in medicine, electronics, for optical devices, and catalysis (Yong et al., 2002, 2003). Especially it is commonly used as catalyst, e.g. in automotive catalytical converters or as catalyst in chemical syntheses. Principally technical recyclability of palladium and other precious metals is considered to be very high, but significant amounts of Pd are released during production processes, consumption and recycling processes. For minimizing loss of Pd and enabling a circular economy a Pd efficient recycling processes while avoiding secondary waste streams of toxic chemicals as well as an efficient recovery of Pd from industrial waste waters are mandatory. The application of Pd(II) reducing microorganisms is an attractive approach that combines the removal of Pd from waste streams, thus minimizing the loss of Pd, with the synthesis of nanocatalysts via bio-reduction and deposition of Pd-nanoparticles on biomass using non-toxic biological means (De Corte et al., 2012). The catalysts itself can be used for the degradation of different recalcitrant pollutants or chemical syntheses.

Different microorganisms have been described that mediate the reduction of Pd(II). In case of the extensively studied anaerobic sulfur-reducing bacteria *Desulfovibrio desulfuricans* and *Geobacter sulfurreducens* and the facultative anaerobic iron-reducing *Shewanella oneidensis* it has been proposed that hydrogenases and cytochrome c3 are involved in bioreduction of Pd(II) and nanoparticle deposition (De Corte et al., 2012; De Windt et al., 2005; Lloyd et al., 1998; Pat-Espadas et al., 2013; Yates et al., 2013). These reactions require H_2 or formate as electron donor. In case of cyanobacteria (e.g. *Plectonema boryanum*, *Anabaena*) it was assumed that the nitrogenase enzyme is responsible for Pd(II) reduction, resulting in the deposition of Pd(0) in the medium, on the cell surface or intracellular. In case of aerobically grown *Escherichia coli* it was postulated that bioreduction of Pd(II) was catalyzed mainly by molybdenum-containing enzyme systems (Foulkes et al., 2016). The majority of the formed Pd(0) particles were formed outside the cell. In other cases Pd(0) formation is not based on enzyme activities. In case of some gram-negative bacteria (e.g. *Paracoccus dentitrificans*, *Pseudomonas putida*, *Cupriavidus necator*) but also gram-positive bacteria (e.g. *Enterobacter faecalis*, (Ha et al., 2016)) Pd(0) nanoparticles were formed in the periplasm, the cell walls, membranes, or intracellular after incubation with formate. In some S-layer carrying gram-positive bacteria Pd(0) were formed after sorption of Pd(II) and subsequent reduction with electron beams or H_2 (Pollmann et al., 2006a; Pollmann et al., 2006b; Wahl et al., 2001). The Pd(0) nanoparticles were deposited in the pores of the S-layer protein. The pore sizes determined the nanoparticle size and Pd(0) localization followed the array structure (Wahl et al., 2001). Binding of Pd(II) complexes as well as nanoparticle formation was extensively studied using different analytical tools proving that carboxylates are involved in complexation (Fahmy et al., 2006; Suhr et al., 2016; Suhr et al., 2014). In all these cases, Pd(0) formation was not restricted to living organisms or entire cells. Experiments with native and dead cells of *E. coli*, *S. oneidensis* and *P. putida*, the protein BSA, and artificial systems composed of amine- and carboxyl functionalized polystyrene particles demonstrated that the presence of amine groups mediates the reduction of Pd(II) bound to cell surfaces (Rotaru et al., 2012). These authors suggested the use of amine-rich biomaterials rather than native cells for Pd-recovery. Consequently, De Corte et al. (2013) replaced the bacteria and used amine-functionalized surfaces as target for the synthesis of Pd(0) nanocatalysts (De Corte et al., 2013). Especially Pd(0) nanoparticles produced on surfaces functionalized with 3-aminopropyltriethoxysilane and chitosan showed a high catalytic activity in comparison to Pd particles formed on *S. oneidensis* cells.

From these findings it can be assumed, that depending on the microorganism several enzymatic and non-enzymatic passive mechanisms may play a role in Pd(II) reduction and influence reduction rate as well as size, localization and distribution of formed Pd(0) nanoparticles.

Pd(II) reducing bacteria have been applied for the removal of Pd(II) from industrial waste streams. This application is a quite attractive alternative to conventional methods, because it requires less toxic chemicals and new products (nano catalysts) are formed as „by-products“. Mabbett et al. (2006) used *D. desulfuricans* in an electro-bioreactor system to recover Pd(II) and synthesis of Pd(0) nanoparticles from a test solution containing a mixture of Pd(II), Pt(IV), and Rh(III) at pH 2.0 (Mabbett et al., 2006). In this approach, up to 79 % of Pd(II), Pt(IV), and Rh(III) were removed and precipitates were formed on cell surfaces and within the periplasm. The formed precipitates were used as catalysts for Cr(VI) reduction in a reactor system. Some studies using *Cupriavidus metallidurans* and *C. necator* gave a proof-of principle for the recovery of Pd from scrap leachates. In case of *D. desulfuricans* high concentrations of Cu(II) that was present in industrial-processing waste stream leachates as well as low pH inhibited hydrogenase activity (Mabbett et al., 2006). Therefore a direct Pd(II) recovery with native cells and reduction to Pd(0) was not possible in this case. However, a „prepalladization“ of the cells via incubation of the biomass with a solution of Pd(II) followed by reduction with H_2 enabled a passive removal of Pd(II) from the leachates (Creamer et al., 2006; Mabbett et al., 2006). Further, the reduction of Au(III) was not inhibited by Cu(II) due to a different reduction mechanism, thus allowing a stepwise separation of the respective metals being present in the leachates (Creamer et al., 2006). Martins et al. (2013) used an anaerobic bacterial consortium composed of *Clostridium*, *Bacteroides*, and *Citrobacter* species derived from a sludge sample from a municipal waste water treatment plant for the removal of Pd(II) from model solutions (Martins et al., 2013). They reached recovery rates of 60 % accompanied by Pd(0) nanoparticle formation. Another group used anaerobic methanogenic granular sludge in a reactor system for the removal of Pd(II) and recovery as bio-Pd(0). The researchers obtained a recovery rate of up to 99 % (Pat-Espadas et al., 2016). In conclusion, principally Pd recovery was possible from waste streams containing pure Pd(II) and/or low metal concentrations. Therefore, such a concept could be especially interesting for the recovery of precious metals from highly diluted waste waters.

In all described cases the formed bio-Pd was catalytically active. Especially the bio-Pd enzymatically produced by *D. desulfuricans* and *S. oneidensis* has shown its applicability for many catalytic reactions. In both cases the produced bio-Pd particles are located on the outer parts of the cells, thus being accessible for surface reactions, have a narrow size distribution and are fastly produced. Generally Bio-Pd was applied to transform a wide range of pollutants, mainly by reduction (Cr (VI), ClO_4^-) (Humphries et al., 2007; Mabbett et al., 2006; Tuo et al., 2013) or dehalogenation (e.g. PCBs, trichloroethylene, pharmaceuticals (Baxter-Plant et al., 2004; De Windt et al., 2005; Hennebel et al., 2010; Hennebel et al., 2009a). In all cases a hydrogen donor (H_2 or formate) was required as the reductive agent. Further, bio-Pd was used as a catalyst for diverse chemical syntheses, e.g. the hydrogenation of organic molecules or for coupling reactions in synthetic organic chemistry (Creamer et al., 2007). A newer approach is the doping of bio-Pd with other metals, e.g. Au, to produce bimetallic catalysts. Such a doping significantly enhanced catalytic activity. This relatively new approach will extend the applicability of metallic bio-catalysts.

Different reactor systems were designed for the application of bio-Pd producing bacteria. Microcosms based on *Clostridium pasteurianum* were used for the remediation of groundwater contaminated with Cr(VI) (Chidambaram et al., 2010). Membrane reactors used for retention of bio-Pd, e.g. reactors based on hollow fiber membranes or plate membrane reactors, were developed for the removal of iodinated contrast media or chlorinated pollutants from ground water. In other concepts bio-Pd was immobilized on membrane surfaces or encapsulated in different polymers or in sol gel matrices or as biofilms on polyurethane (Hennebel et al., 2009b; Humphries et al., 2006; Pollmann et al., 2006a; Quan et al., 2015b; Yong et al., 2007). These materials were used for the removal of pollutants or for the construction of microbial fuel cells. However, catalytic activity was reduced in most cases in comparison to suspensions of bio-Pd. Besides the native bacteria, also the bacterial S-layer proteins have been proven to be an appropriate support for catalytic active bio-Pd. The self-assembling properties of S-layer proteins enable the coating and functionalization of various technical surfaces, thus providing the basis for the construction of catalytic active thin layers (Weinert et al., 2015a). S-layer based bio-Pd has been used for the targeted regular deposition of Pd(0) nanoparticles on diverse technical surfaces (Mertig et al., 1999; Pompe et al., 1996; Suhr et al., 2014). These coatings were used for the electro less deposition of metals on surfaces or for chemical syntheses (Creamer et al., 2007; Dieluweit et al., 2005; Huttl et al., 2009).

Figure 3. Pd(0) nanoparticles supported by S-layers visualized using a helium ion microscope (Hlawacek et al., 2014). The insert shows the Fourier spectrum of the indicated area showing the regular square arrangement of the Pd(0) nanoparticles. The distance between the Pd(0) nanoparticles, as measured from the Fast Fourier Transform (FFT), is 12 nm.

Besides catalytically applications, bio-Pd has been applied in microbial fuel cells, e.g. proton-exchange membrane fuel cells, for the generation of energy. In these approaches, biologically produced Pd(0) particles were deposited onto the anode, e.g. carbon papers, of the fuel cells and used for energy generation (Quan et al., 2015a; Yong et al., 2010; Yong et al., 2009). The thus modified anode possessed both electro oxidation and biodegradation capability (Quan et al., 2015a).

In recent research different bacteria (*S. oneidensis*, *C. metallidurans*, *G. metallireducens*, *P. stutzeri*, *Bacillus toyonensis*) have been applied for the recovery of different platinum complexes at pH 2 in the presence of H_2 as electron donor (Maes et al., 2017). Pt particles were precipitated intra- and extracellularly. Further, halophilic bacteria were used for the recovery of Pt complexes from saline solutions (Maes et al., 2016). Also Au(III) could be removed from electronic scrap leachates (Creamer et al., 2006). The anaerobic bacterium *Shewanella algae* was used for the bioreductive production of Au, Pt, and Pd particles (Konishi et al., 2006; Konishi et al., 2007a; Konishi et al., 2007b; Tamaoki et al., 2010). Ito et al. (Ito et al., 2016) genetically constructed yeasts displaying dehydrogenase on their cell surface and used it for the reduction of Pt(II) and precipitation of Pt(0). These examples demonstrated that the concept of Pd recovery can be transferred to other precious metals at various conditions.

6. Conclusions and future prospects

In conclusion, there are different biotechnological concepts aiming the recovery of metals from various waste materials. These concepts are based on natural mechanisms of organisms or biomolecules interacting with metals, such as transformation, complexation, or adsorption. Currently only few of these developments are commercialized. Major limitations are high costs in comparison to conventional methods and low element selectivity. However, increasing environmental consciousness as well as limitations of classical methods especially in case of complex materials with low metal content will boost novel developments. The present article is focusing on the fields of bioleaching, biosorption, bioflotation, and bioreduction and gives an overview on promising developments.

Bioleaching has been intensively studied for sulfidic ores using chemolithoautotrophic bacteria and commercial applications have been established (Rawlings, 2002; Vera et al., 2013). Most of these processes aim the recovery of copper from sulfidic ores by using irrigation-type processes (e.g. heap leaching). Other stirred tank processes are focused on the pretreatment of ores via a biooxidative process for the recovery of gold from recalcitrant arsenopyrite (Rawlings, 2002).

More recent bioleaching approaches concentrate on non-sulfidic materials, including diverse waste materials, and explore the potential of a larger variety of natural microorganisms to extract metals at a broad range of conditions. Whereas numerous studies investigated the release of elements such as Cu, Au, only few studies concentrate on more rare elements such as In, Ga, or REE. In view of the large number of elements, the growing complexity of technical poducts and the need of recycling it can be expected that future studies will include a broader range of materials and extend the application range for bioleaching technologies. Further, the combination with chemical methods, sequential extraction as well as sophisticated process design (e.g. (Jadhav et al., 2016; Wang et al., 2016; Zhao et al., 2008)) will enhance bioleaching efficiencies and reduce costs.

In case of biosorption, many efforts have been done ranging from the application of different biomasses, constructing biosorptive composites, design of metal binding molecules, and use of different metabolic microbial processes. These approaches were used especially for the removal of toxic elements from waters. There are some reports on the removal of precious metals but relatively few studies describing the recovery of other value metals, e.g. REE, Ga, or precious metals (Gascoyne et al., 1991a; Moriwaki and Yamamoto, 2013; Park et al., 2017; Pollmann et al., 2006b; Yunus and Tsai, 2015). It can be assumed that many technologies developed for heavy metal removal can be transferred to other elements. Thus a high potential for future applications in recycling technologies can be expected. Further modern biotechnologies such as the specific design of biomolecules like proteins or peptides, surface functionalization and design of composites, but also drawing on natural resources such as an incountable number of natural products, will open new perspectives and push developments in this research field.

Bioreduction has been mainly applied for the removal of Pd(II) from industrial waste streams (Mabbett et al., 2006). However, the methods can be principally also applied for other precious metals such as Pt or Au (Creamer et al., 2006; Konishi et al., 2007a; Konishi et al., 2007b; Maes et al., 2016). The nanoparticles that are formed during the reduction can be applied as nano catalysts, e.g. for the transformation of pollutants or chemical syntheses (De Corte et al., 2012). Recent research activities investigate a broader range of bacteria, e.g. by using halophilic bacteria at different process conditions, apply genetic methods, or use novel bioreactor systems (Ito et al., 2016; Maes et al., 2016; Maes et al., 2017; Pat-Espadas et al., 2016).

Bioflotation is a highly interdisciplinary approach that enable the separation of particles by exploiting the natural interactions of biocompounds with surfaces. The biocompounds (mostly cells or metabolic compounds) are added as reagent in flotation processes. Most current studies used microbial cells or EPS for mineral separation (Behera and Mulaba-Bafubiandi, 2017; Govender and Gericke, 2011). These compounds have a complex composition, consisting on various functional groups and molecules that interact with particle surfaces and alter their properties. Other researchers used more defined biomolecules such as DNA or biosurfactants (Khoshdast et al., 2012b; Vasanthakumar et al., 2012). Very recent publications suggested the use of phages, peptides, or proteins for particle separation (Curtis et al., 2009; Shen et al., 2017; Vreuls et al., 2011). Generally, this field is little studied and not yet applied to recycling methodologies. Therefore many future developments can be expected.

Other approaches such as bioprecipitation, biomineralization, and microbial electrochemistry are not considered in detail in the present article (Fang et al., 2011; Modin and Aulenta, 2017; Modin et al., 2012). These methods are in particular promising for the recovery of metals from highly diluted solutions.

In summary especially the combination of different scientific disciplines and technological approaches, the integration of novel post-genomic biotechnological methods as well as the consideration of existing, yet unexplored natural resources may drive innovations in metal recovery.

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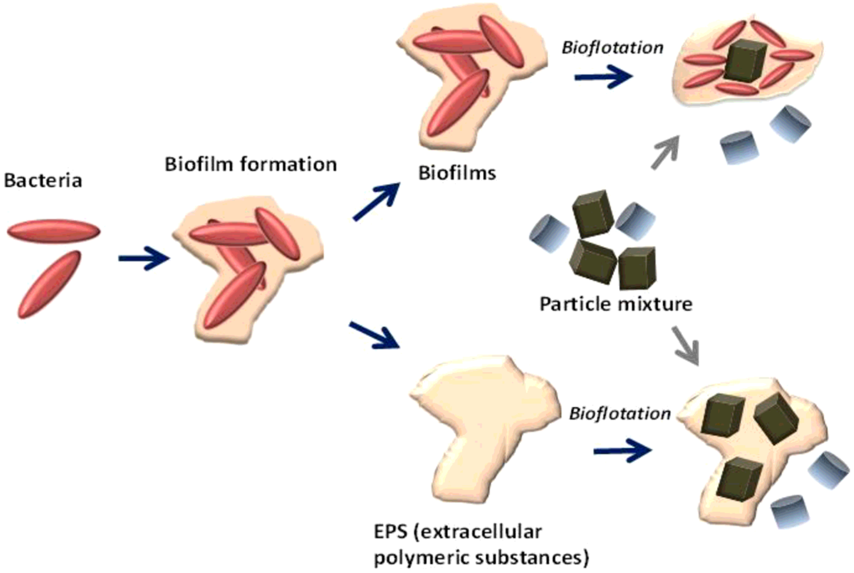
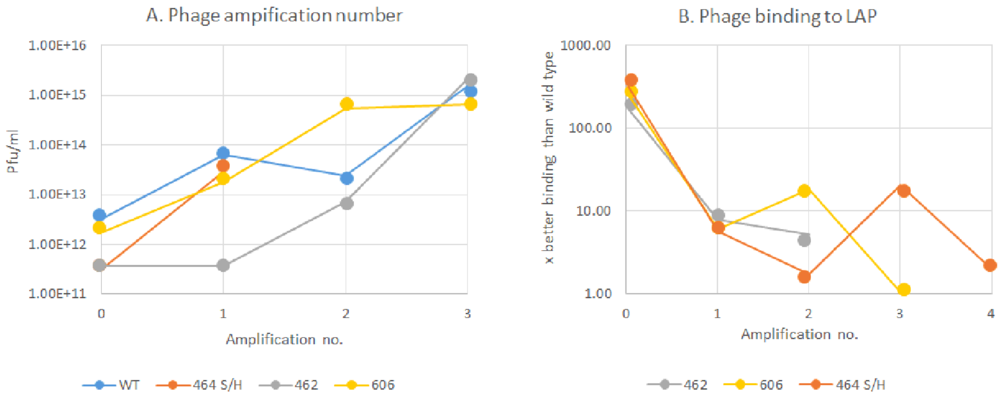

Bacteria
Biofilm formation
Biofilms
Bioflotation
Particle mixture
Bioflotation
EPS (extracellular polymeric substances)

Fig. 1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1.00E+14
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1.00E+11
Pfu/ml
0
1
2
3
Amplification no.
WT
464 S/H
462
606
B. Phage binding to LAP
1000.00
100.00
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x better binding than wild type
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Fig. 2

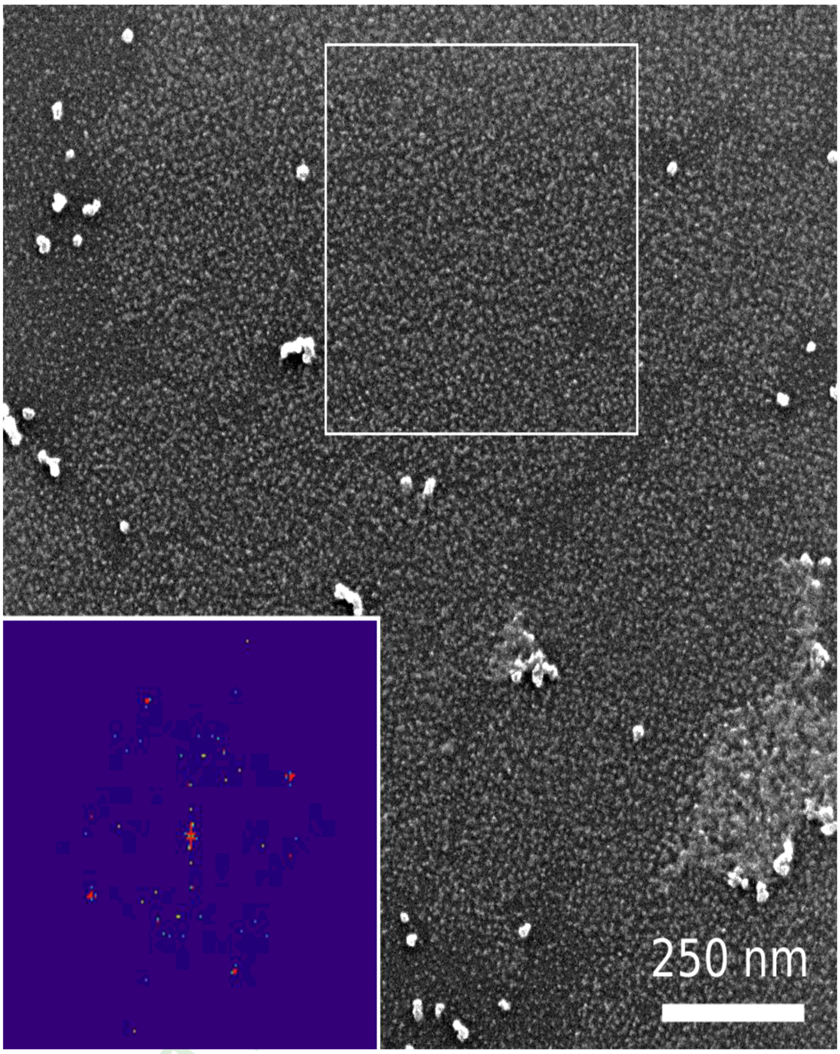

Fig. 3