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Removal of nickel(II) from industrial wastewater using selected methods: a review

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Due to the increasing problem resulting from environmental pollution with heavy metals, great emphasis is placed on the development of removal methods of these pollutants from the environment. This study presents a literature review on the methods for the removal of nickel ions from aqueous solutions such as sorption, especially using low-cost sorbents which are very popular in 21st century, electrochemical processes and membrane techniques. It is often impossible to use a single technique for efficient removal of heavy metals from wastewater as the process depends on many factors, such as wastewater composition, pH, temperature and many others. The aim of this review is to present some selected removal techniques of nickel(II) from wastewater from the point of view of their efficiency and applicability.

Keywords: nickel; nickel removal; wastewater; adsorption; electrocoagulation; membrane techniques

1. INTRODUCTION

Nickel is a silvery-white metal with a golden sheen, which is hard and malleable, and belongs to the group of transition metals. Next to iron, oxygen, silicon and magnesium, it is the most common element in the environment. It exhibits ferromagnetic properties and it corrodes slowly in spite of its susceptibility to oxidation. Due to its considerable gloss, it is used in ornamentation and ceramics. However, nickel is used mainly in the production of everyday objects. Nickel is a common allergen which can cause contact dermatitis. This is the symptom of the most commonly perceived danger from this metal. Nickel is of great biological significance and is classified, on the one hand, as a trace element and, on the other hand, as a substance which is particularly harmful to the aquatic environment (Dz.U. 2019 poz. 1220).

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In the industry, nickel is mainly used as a coating for the less noble iron and steel in order to increase the strength and corrosion resistance of these materials. Such steels are used e.g. in the automotive industry.

The nickel crystal lattice has the ability to absorb hydrogen atoms. The highly fragmented metal, known as Raney nickel (the so-called backbone catalyst), can hold approximately 17 times more hydrogen than its volume. Due to this property, nickel is used in reduction processes, including the hydrogenation (chemical hardening) of fats.

Nickel is also a component of nickel-cadmium batteries. It is expected that nickel will soon be the leading metal in battery production processes, both in pure metal form (e.g. electroplating) and in the form of alloys, including water-absorbing alloys, which are an element of many modern energy storage devices. It should be emphasised that the turn of 20^{th} and 21^{st} centuries was a time of great changes in the area of chemical energy storage. The last decade has seen rapid market growth associated with "electromobility", the share of which in 2000 was negligible. The growth dynamics of the chemical energy storage segment is illustrated in Figure 1 (Pillot, 2014).

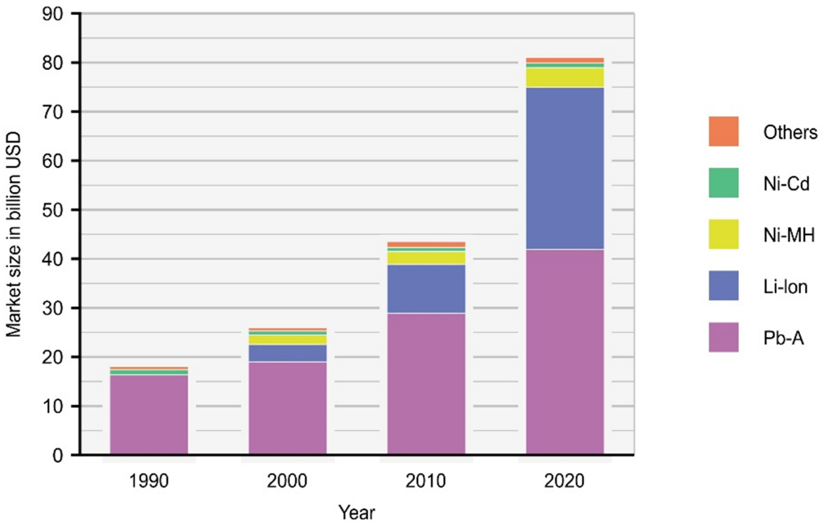

Fig. 1. Global market for secondary cells (Pillot, 2014)

The batteries currently dominating the market are lead–acid (Pb–A) and lithium–ion (Li–Ion) systems. However, nickel–metal hydride (Ni–MH) systems come next in terms of market demand (Lota et al., 2020). The widest application of nickel is in the production of alloys, including so-called superalloys (alloys based on nickel, iron and cobalt) or nickel–copper alloys for coin production. Superalloys are used in the energy industry due to their heat resistance and low material creep at high temperatures. Nickel is also sometimes used in the manufacture of spectacle frames and jewellery.

2. METHODS OF REMOVING NICKEL FROM INDUSTRIAL WASTEWATER

Wastewater generated during production processes, e.g. galvanic wastewater or steel leaching solutions, as well as wastewater from metal surface treatment using organic solvents, are classified as particularly hazardous and burdensome for the environment due to high content of heavy metals (within the range of 10 to 1000 mg/dm^3). The presence of pollutants, e.g. in galvanising plant effluents, influences the change of water pH, the amount of dissolved oxygen and the organoleptic properties. Unfortunately, a high content of heavy metals also negatively affects the process of water self-purification, therefore appropriate water and sewage management are important elements of environmental protection (Milh et al., 2020; Ramírez

Calderón et al., 2020). Special attention should be paid to new prototype methods of recovery and use of heavy metals. Wastewater management planning should take into account the fact that wastewater after heavy metal recovery ultimately ends up in natural waters. If the wastewater is discharged first to a sewer and then to a treatment plant in a collective system, the wastewater must meet the parameters specified in the Announcement of the Minister of Infrastructure and Construction of 2016 (Dz.U. 2016 poz. 1757). If the wastewater is discharged into waters, it must meet the requirements of the Regulation of the Minister of Maritime Affairs and Inland Navigation of 2019 (Dz. U. 2019 poz. 1311). The limit values for nickel concentrations are 1 mg Ni/dm^3 if the recipient is sewerage and 0.5 mg Ni/dm^3 if the recipient is the aquatic environment, respectively. The emission standards for nickel are being tightened with the development of technology. For example, the 2020 updated reference document (BAT) for surface treatment using organic solvents (Chronopoulos et al., 2020) indicates a reference value for nickel emissions in wastewater in the concentration range of 0.05 to 0.4 mg Ni/dm^3, irrespective of whether the discharge is direct or indirect to a receiving water body. Therefore, there is a need to develop and introduce methods for the recovery of nickel as a raw material and advanced techniques for the removal of nickel from waste streams discharged into the environment. Such an approach is perfectly in line with the new economic model – the circular economy. The possibility of returning raw materials to the process reduces the costs of its implementation, while limiting the release of harmful components into the environment. This solution allows to close a product's life cycle and move from a linear economy model (acquisition of raw material – production – use – disposal of waste) to a circular model (production – use – reuse of waste as a raw material in the next production cycle). The recovery of nickel from waste solutions as a valuable raw material for the production of, for example, water-absorbing materials, is part of this solution.

Important groups of methods for the removal of nickel from aqueous solutions include sorption processes based on low-cost sorbents, electrochemical processes and membrane techniques.

Sorption, considered as an environmentally friendly operation, deserves special attention as a method for removing harmful nickel from aqueous solutions. The classical adsorbents are activated carbons used, among others, for remediation of inorganic metal contaminants. Activated carbons (AC) are characterised by high adsorption capacity, high specific surface area, regenerability and specific surface reactivity. The adsorption capacity of example carbon adsorbents for remediation of Ni(II) ions is presented in Table 1.

Table 1. Adsorbents for Ni(II) ion remediation (Bohli et al., 2015)

Adsorbent	Removal efficiency [mg/g]	Thermodynamics of reactions	pH
Charcoal	96.0	Endothermic	7.5
Activated charcoal	99.0	Endothermic	7.5
Walnut shell carbon	15.34	Endothermic	6.0
Citrus seed carbon	35.54	Spontaneous and exothermic	6.5
Lignite	13.0	Exothermic	4.5–5.5
Lignite Beysehir	12.0	Exothermic	4.5–5.5

Due to the high environmental costs of using classical sorbents, so-called low-cost sorbents are currently being sought. Generally, low-cost sorbents can be divided into five groups (De Gisi et al., 2016): (i) agricultural and household waste, (ii) industrial by-products, (iii) sludge, (iv) sea materials, (v) soil and ore materials, and (vi) novel low-cost adsorbents. In this group, agricultural wastes are used as adsorbents for the removal of nickel ions. These materials are of broad interest due to their low cost and high availability. The main components of agricultural waste are cellulose and lignin, which contain various functional groups, i.e. hydroxyl, carbonyl, carboxyl and ether bonds. These groups have the ability to strongly interact with heavy metals and use electron pair to form complexes (Bohli et al., 2015; Ewecharoen et al., 2008). Examples of the use of agricultural materials for the removal of Cu(II), Ni(II) and Pb(II) ions are presented in Table 2.

Table 2. Comparison of the removal efficiency of selected metal ions (mg/g) using activated carbons with those obtained from different plant materials (Bohli et al., 2015)

Adsorbent	Conditions	Cu(II)	Ni(II)	Pb(II)
Granular AC	pH 5	15.58	–	15.58
Powdered AC	–	26.9	–	26.90
Apricot kernels	pH 6.5, 25 °C	24.08	26.9	22.84
Carbon nanotubes	pH 5	28.6	–	96.97
Bamboo charcoal	pH 5, 23 °C	7.4	6.46	–
Activated carbons from olive stones – chemical activation (COSAC)	pH 5, 30 °C	17.78	24.07	148.8

The maximum adsorption capacity of different adsorbents made from agricultural waste is summarized in Table 3.

Table 3. Maximum nickel(II) adsorption capacity of different adsorbents obtained from agricultural waste (Bohli et al., 2015; Charazińska et al., 2022; Ewecharoen et al., 2008; Rico et al., 2018)

Adsorbent	Removal efficiency [mg/g]	Thermodynamics of reactions	pH
Barley straw	35.8	–	4.5
Rice bran treated with hydrochloric acid	149.4	–	6.0
Rice bran protonated	102.0	–	6.0
Waste pomegranate seeds	52.00	Endothermic	6.0
Cashew nut shells	20.0	Spontaneous and exothermic	5.0
Waste acorns of Quercus ithaburencis (WAQI)	9.42	Endothermic	5.0
Citrus peel (CLPC)	38.46	Spontaneous and exothermic	6.0
Sugar cane bagasse	16.31	–	5.0

Stevens and Batlokwa (2017) have published data regarding the use of eggshell biomass for nickel and cobalt removal. The pH value of the aqueous solution is very important in sorption processes as it affects the solubility of metal ions, the concentration of counter ions in the functional groups of the adsorbent and the degree of ionisation of the sorbate during the reaction. As the number of sorbent particles surrounding the metal ion increased, the metal removal efficiency increased.

Industrial solid wastes, i.e. fly ash, blast furnace slag, red sludge, waste sludge and bio-particles were also used for nickel ion removal (Schlögl, et al., 2022). Fly ash generated in thermal power plants is also used as an adsorbent.

Nettle ash was also used to remove Ni(II) from aqueous solutions (Mousavi and Seyedi, 2011). The adsorption capacity of Ni(II) ions was 192.3 mg/g. The adsorption efficiency was affected by various parameters, i.e. pH, temperature, initial concentration and contact time. The results obtained were consistent with the Langmuir isotherm model and kinetic studies showed that the adsorption kinetics of Ni(II) was consistent with pseudo-second order models. Using blast furnace slag, a Ni(II) removal capacity of 55.75 mg/g at pH 5.0 was obtained. The adsorption results were described by the Freundlich isotherm and it was also found that the adsorption of Ni(II) on the slag was exothermic. [36]Van Hullebusch et al. (2005) used anaerobic granular sludge as a low-cost adsorbent for Ni(II) removal from industrial wastewater. The adsorption capacity of Ni(II) ions was 13.3 mg/g at pH 7.2. The experimental results are well described by the Langmuir isotherm model and consistent with a pseudo-second order kinetic model.

The maximum adsorption capacity of Ni(II) ions using adsorbents obtained from industrial waste is presented in Table 4.

Table 4. Maximum adsorption capacity of adsorbents derived from industrial waste for remediation of Ni(II) ions (Stevens and Batlokwa, 2017)

Adsorbent	Removal efficiency [mg/g]	Thermodynamics of reactions	pH
Red mud	13.69	Endothermic	5.0
Nettle ash	192.3	–	6.0
Water-cooled blast furnace slag	12.66	Endothermic	6.0
Cleaned slurry (steel industry)	14.30	Endothermic	5.0
Anaerobic pelletised sludge	13.30	–	7.2

In 2020, a team of Chinese researchers (He et al., 2020) proposed the possibility of using zeolites derived from coal fly ash for highly efficient removal of Ni(II) ions. The report proposes the preparation as a sorbent – a zeolite derived from coal fly ash modified by sintering to remove carbon, then acidified to remove iron, activated with alkali, and then hydrothermally crystallised. The process parameters, mechanism and kinetics of Ni(II) ion removal were studied. The results indicate that Ni(II) removal reached up to 94%, the adsorption capacity was 47 mg/g with an initial content of 100 mg/dm^3 Ni(II).

Activated zeolite in the form of Na-mordenite was used as an adsorbent to remove nickel(II) from aqueous solution, and exhibited higher values of specific surface area, total pore volume, and average pore size compared to mordenite (Mehdi et al., 2022). The maximum adsorption capacity predicted by the Langmuir isotherm was found to be 36.79 mg/g at 60 °C, while the experimental results of kinetic and equilibrium studies were expressed the best by the Avrami model and the Redlich Peterson isotherm.

For the removal of nickel from aqueous solutions, natural mineral sorbents are also used and their adsorption capacity towards nickel is presented in Table 5.

Nanotechnology is also used to remove toxic pollutants from aqueous solutions, i.e. lead, chromium, arsenic, nickel, etc. The special properties of nanomaterials and the sorption properties of some of them are used in water and wastewater technology. Generally, nanoadsorbents are classified in some types (Vishnu et al., 2021): (i) metallic nanoparticles, (ii) carbonaceous nanomaterials, (iii) silica nanomaterials,

Table 5. Adsorption capacity of the selected mineral sorbents (Kumar et al., 2019; Mehdi et al., 2022)

Adsorbent	Removal efficiency [mg/g]	Thermodynamics of reactions	pH
Vermiculite	25.4	Endothermic	6.0
Montmorillonite	46.0	–	6.0
Clay	97.0	Endothermic rection	7.5
Natural bentonite	50.0	Spontaneous and endothermic	8.0
Na–bentonite	13.96	–	6-8
Na–mordenite	36.79 at 60 °C	Spontaneous and endothermic	5.30

(iv) nanofibers and nanoclays, (v) polymer composites, (vi) aerogels. Nanomaterials are currently used in the form of nanofibres, nanoparticles, dendrimers and nanotubes (Li et al., 2018; Noman et al., 2022). Modern nanostructured materials are robust and stable.

For example, enhanced efficiency for nickel(II) removal from wastewater using nanocellulose (NC) and nanoscale zero-valent iron (nZVI) composite (NC–nZVI) was confirmed (Song et al., 2022). The anchoring of nZVI at the functional sites of biomass derived NC was carried out by one-step liquid-phase reduction method. Cellulose nanocrystals–nZVI sorbed over 80% of nickel(II) in the pH range from 3.0 to 5.0 in the presence of varied ions (NO^{3-}, Cl^- and Ca^{2+}), indicating potential for practical applicability.

Another type of nonosorbents, i.e. γ-alumina nanoadsorbent with high or low specific area (342.42 or 236.56 m^2/g, respectively), was produced by solvothermal method (Kesthkar et al., 2021). The nanoadsorbent with high and low area removed 99.6 and 96.9% of nickel(II), respectively, from synthetic wastewater under the optimum conditions, i.e. 40 °C, 2 g of adsorbent, pH 4.0, initial concentration of nickel(II) 25 mg/dm^3, 60 min contact time.

Electrochemical processes, mainly electrocoagulation, can also be used to remove Ni(II) ions. [1]Akbal et al. (2011) studied the elimination of copper, chromium and nickel from galvanic wastewater using iron and aluminium electrodes. The effects of pH, current density, electrode material and conductivity on nickel removal efficiency were also investigated. The results show that electrocoagulation with a pair of Fe/Al electrodes was very effective. A 100% removal efficiency of copper, chromium and nickel was achieved in 20 minutes, at pH 3.0, and a current density of 10 mA/cm^2.

[35]Un et al. (2015) studied the elimination of heavy metals Cd, Cu and Ni by electrocoagulation from industrial wastewater. They observed that nickel removal efficiency was affected by pH and current density. The removal efficiencies of Cd, Cu and Ni were equal to 99.78, 98.90 and 99.98% at a current density of 30 mA/cm^2 and pH 7, respectively.

[3]Arabameri et al. (2022) found that a transition from conventional direct current (DC) to alternating current (AC) waveform electrocoagulation led to a more than 16% reduction in energy consumption and approximately 47% depletion in electrode consumption during the removal of nickel(II) from synthetic wastewater.

Electrocoagulation is also used to remove nickel(II) from wastewater (Kumar et al., 2019) During coagulation, aluminium ions from the electrodes react with Ni(II) ions, resulting in precipitation. The efficiency of electrocoagulation is influenced by various parameters, i.e. the applied current density, pH and initial ion concentration. The removal efficiency of nickel ions with this method ranges from 88.60 to 99.5%. The use of electrocoagulation for simultaneous elimination of ions of various heavy metals, i.e. mercury, lead and nickel, is also described. It was shown that the maximum efficiency of heavy metal removal was obtained at pH 7.0, with a current density of 0.15 A/dm^2. A list of electrochemical technologies used to treat Ni(II) ions is given in Table 6.

Table 6. List of electrochemical technologies for Ni(II) ion treatment (Kumar et al., 2019)

Technique	Cathode	Anode	Removal efficiency [%]	pH
Electroflotation	Aluminium	Aluminium	99	8.0
Electrocoagulation	Aluminium Stainless steel	Aluminium Stainless steel	–	4.5–7.5
Electrodeionisation	Platinum Platinum Titanium Platinum	Platinum Graphite powder Platinum powder	100	4.0
Electrodialysis	Pt oxide-coated Titanium	Stainless steel	69	8.0

Membrane techniques are an important group of methods for removing nickel(II) from aqueous solutions, especially in hybrid solutions. In 2018, Chinese researchers proposed a PAA/ZIF-8/PVDF hybrid ultrafiltration membrane for nickel(II) removal from high-salinity wastewater. The membrane was fabricated by immobilising zeolite imidazole backbone-8 (ZIF-8) particles on the surface of a trimesoyl chloride (TMC)-modified polyvinylidene fluoride (PVDF) membrane under the protection of a polyacrylic acid (PAA) layer. The obtained PAA/ZIF-8/PVDF membrane showed relatively high uptake of nickel(II) ions (219.09 mg/g) from high salinity wastewater due to adsorption of metal ions on the membrane. Spectroscopic studies have shown that the removal of Ni(II) from solutions is mainly due to the specific interaction between Ni(II) and hydroxyl groups in the ZIF-8 skeletons and carboxyl groups in the PAA layer (Li et al., 2018).

Among the membrane techniques used to remove heavy metals from industrial wastewater, the use of liquid membranes (an organic phase is a semipermeable barrier between two liquid or gas phases) for selective removal of heavy metal ions should be mentioned. Generally, liquid membranes are classified according to a module configuration (BLM – bulk liquid membranes, ELM – emulsion liquid membranes, SLM – supported liquid membranes), transport mechanism, application, type of carrier, type of membrane support (Kislik, 2010). Liquid membrane-based extraction processes have become widely investigated and applied for removal of various contaminants, especially metal ions, from waste solutions. Membrane techniques are continuously improved and developed, are considered environmentally friendly and can be used in processes in which commonly known methods are not efficient or selective (Rzelewska-Piekut and Regel-Rosocka, 2021).

Highly efficient and selective membrane separation of copper from nickel from ammoniacal solutions using flat-sheet supported liquid membranes (FSSLM) with mixtures of metal carriers (Acorga M5640 and bis(2-ethylhexyl)sulfoxide) was proposed by [9]Duan et al. (2020). FSSLM appeared to facilitate transport of copper(II) and separate copper(II) efficiently from nickel(II) (separation factor of 26.3). Furthermore, stability of the membrane was confirmed in at least ten separation cycle runs.

To increase the driving force and overcome the diffusion limitations of liquid membrane transport, some additional operations can follow the membrane process. For example, [24]Mondal et al. (2019) proposed an integrated liquid membrane – electrowinning (LM–EW) system for transport of metal ions (e.g. nickel and zinc from wastewater) stripped from the LM to the receiving phase. Electrodeposition of metals allows to keep the driving force constant, resulting in a high effectiveness of the separation processes, and to obtain valuable metallic products.

The results of laboratory tests carried out by many scientists indicate the possibility of using liquid membranes to reduce the content of metal ions in industrial wastewater to a level which allows to discharge it into the sewage system. Moreover, the metal ions separated in this manner can be returned for reuse in the technological process, which does not generate waste, in contrast to e.g. the classical method of chemical precipitation, and is beneficial both from an ecological and economic point of view (Kończyk, 2019).

3. COMPARISON OF THE PRESENTED METHODS OF NICKEL(II) REMOVAL

Each of the presented methods of nickel(II) removal has some advantages that make it important for practical applications and drawbacks which should be outweighed by such merits as selective metal separation from low concentrated wastewater or from deposits of unconventional composition, high purity of the final product, well-known mechanisms of reaction and mass transport, wide range of technical and technological solutions, etc. The comparison of merits and demerits of the nickel(II) removal methods presented in this review is shown in Table 7.

Table 7. The comparison of merits and demerits of the selected methods of nickel(II) removal from wastewater

Method	Merits	Demerits	Ref.
Adsorption on activated carbons (AC)	• Simplicity of the method • Convenient and safe operations • Technical applicability for a wide variety of substances • Good efficiency of adsorption • Well-known and widely studied adsorbent	• Increase in volume of diluted solutions after elution • Not for concentrated metal solutions • Nonselective and highly sensitive to solution pH • High cost of AC regeneration	Gupta et al. 2009; Sadegh and Ali, 2018
Adsorption on low-cost sorbents (LCA)	• Low-cost of sorbent production (cheaper than AC) • Reducing of waste/biomass by-products • Abundance of raw materials • Low regeneration cost • Circular economy approach	• Low surface area of many LCA • Lower efficiency of adsorption than AC	Bohli et al., 2015; Gupta et al. 2009
Adsorption on nanosorbents (NS)	• Enhanced adsorption capacity • Enhanced surface to volume ratio of NS • Possibility to tailor the surface properties • Reusability and easy recovery	• Needs more R&D • Only laboratory scale investigation • Technical challenges: scale-up, system setup • Environmental concerns • High cost of NS manufacturing • Only a few commercial products available in the market	Bhatnagar and Sillanpää, 2014; Vishnu et al., 2021

Table 7 continued

Method	Merits	Demerits	Ref.
Electrocoagullation	• High removal efficiency • No need for supplementary chemicals • Environmental compatibility • Ease of operation	• High consumption of energy • High cost of electrode materials	Arabameri et al., 2022
Liquid membrane extraction	• Variety of LM types appropriate for different applications • Small space requirement • Avoiding the necessity of phase separation • Use of highly selective carriers of metal ions	• Lower yield than in case of liquid-liquid extraction • Need for assembly of many membrane modules • Problems with membrane stability	Alguacil et al., 2011; Rzelewska-Piekut and Regel-Rosocka, 2021

Depending on source and composition of the wastewater, a suitable treatment method should be selected. For example, the high cost of AC regeneration resulting from energy-consuming thermal processes can be avoided when low-cost adsorbents are used because the heat treatment of these sorbents is simpler and cheaper (De Gisi et al., 2016). However, some proposed methods (e.g. adsorption on NS) are in the early stages of the development (laboratory scale), and still many issues need attention, such as research with real industrial effluents, investigation of material regeneration, and studies in continuous flow systems.

4. SUMMARY

The increase in environmental awareness, as well as the changing needs of the market and the simultaneous reduction in the abundance of natural raw materials, have made the recovery of metals from waste solutions an important research and practical issue in recent years. Moreover, due to the increasing amount of various types of waste and used products, ways of their management and utilization are being sought, which not only protect the natural deposits of raw materials, but also contribute to the reduction of the amount of accumulated waste and promote the implementation of solutions supporting the principle of a circular economy.

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