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SUSTAINABLE MANAGEMENT, TREATMENT AND RECOVERY OF WASTE IN AN EUROPEAN CONTEXT: URBAN MINING AND THE CIRCULAR ECONOMY

BY

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Abstract. The paper provides an overview of waste management alternatives in the European framework of the circular economy model, with emphasis on the use of waste as secondary sources for the recovery of raw materials and materials, especially critical raw materials and highlighting the importance of economic and environmental impact of this approach. In this context, the following were taken into account: analysis of European urban waste management practices; highlighting the relevance of waste recovery in the context of the circular economy; recovery of critical raw materials (CRM) from specific waste, as secondary sources of raw materials. In the last part of the paper, a series of methods for the recovery of secondary and critical raw materials from waste were highlighted, analyzing the case of recovery of cobalt from secondary resources, such as waste lithium batteries, as important for lithium and a few other critical materials, as well.

Keywords: critical raw materials, recovery, recycling, secondary resources, waste.

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1. Introduction

Waste is currently the best indicator available to describe the general development of the society, since this is associated with waste generation and treatment in European countries. All countries collect data on municipal waste, but data coverage for different wastes, for example total waste or household waste is quite limited. Municipal waste constitutes about 10% of the total waste generated, but due to its complex nature and distribution from many waste producers, the sustainable management of this waste is relatively complicated (EEA, 2018; Ghinea *et al.*, 2022). Current trends in the approaches for waste management must take into account two main issues: a small amount of waste; avoidance of environmental pollution. In this context the waste management hierarchy asks for prevention of waste generation; waste treatment at the source of their generation; promoting recycling by any means, reuse, compost production; use of final disposal methods (landfill, incineration) for waste that cannot be recovered (EC Directive 98, 2008).

The amount of solid waste generated by population and commercial and industrial activities has reached such levels that it creates problems for an accurate land management system. These problems cannot be solved by arranging new spaces for landfills or by installing new incinerators that are expensive and raise complex problems regarding protection systems (Angelescu *et al.*, 2003). We must appreciate some household waste (paper-cardboard, plastic, glass, metallic materials, organic matter, etc.) as sources of secondary raw materials. Figure 1 shows the trend in waste generating and management in the European Union (EU) from 1995 to 2018 (Eurostat, 2019). A very distinctive trend in waste disposal lessening can be observed, since countries continue to select alternatives for sustainable waste treatment and recovery of materials and energy.

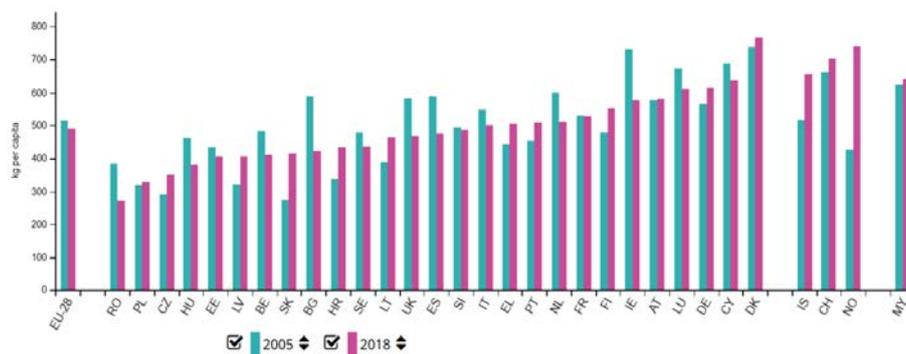


Fig. 1 - Municipal waste generated in Europe, compared in 2005 and 2018 (kg per capita) (Eurostat, 2019).

Urban waste is increasingly considered as secondary sources for raw materials and energy, insufficiently exploited by recycling, reuse in efficient systems. This approach also has an ecological side, because by reducing the amount of waste, a series of problems concerning the quality of the environment and human health can be solved.

2. Waste management hierarchy

Over the last 30 years, European policy efforts have led to a number of environmental action plans, directives, reports and a legislative framework aimed at reducing the negative impact of waste on the environment and health and improving the energy efficiency of resources at European level (EC, 2010). For example, the EC Directive 98 (2008) (Waste Framework Directive) established a five-step "waste hierarchy", "producer responsibility" and the "polluter pays principle". The waste management framework places priorities for waste management with the preferred waste prevention option, followed by reuse and recycling, devoted to close the life cycle of a product. At the bottom of the hierarchy is the recovery of energy from waste, the option of storing landfills is the least desired. Therefore, in order to improve waste management, actions are prioritized according to the "waste hierarchy" (Fig. 2).

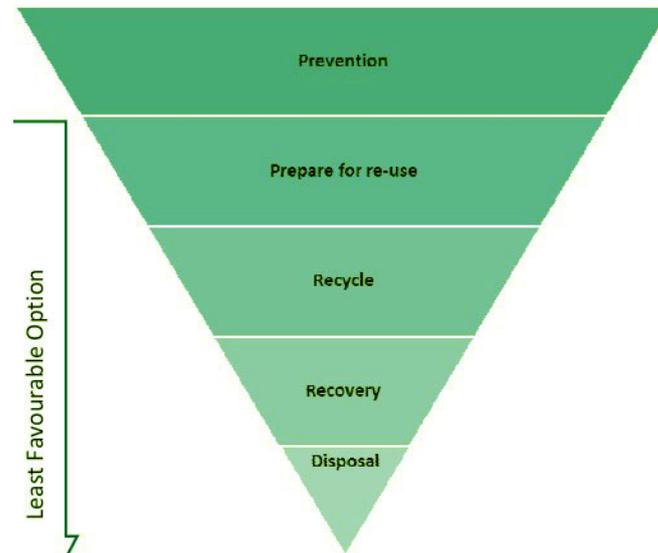


Fig. 2 – Waste management hierarchy.

Resource depletion is correlated with the major indirect impact on the environment, associated with the extraction and processing of resources, so as to reward the resources which do not take part any more in the economy circuit. As

stated in the EC Directive 98 (2008), all the alternative needs to be considered for ensuring waste management hierarchy in the view of its full implementation, this meaning waste prevention and reuse, recycling, recovery, disposal (Fig. 2) in order to avoid significant impacts on the environment. In this context, waste disposal can generate direct environmental impacts on land use, resources depletion, methane and other greenhouse gas emissions with consequences on global warming. In the case of waste depositing, eutrophication and ecotoxicity due to nutrients existing in leachate can manifest as impacts. Depletion of resources, acidification and ecotoxicity effects of air emissions can be produced in case of waste incineration (Everett, 2012).

3. Waste and the circular economy

3.1. General trends in European waste management

Although the planet offers various natural resources, their use in unsustainable production and consumption systems induces significant negative economic and ecological impacts, at national level and in a cross-border context. The intensity of these impacts depends on the type and quantities of raw materials taken from natural resources, the natural environment from which they are extracted and how they are managed (EEA, 2019; Slorach *et al.*, 2019). Although the extensive use of raw materials contributes to the economic development of countries, regions, continents, the downside of this context is the generation of waste through the incorrect use of these resources, the unsustainable functioning of the economic system, but also population growth. Although the solution to the problem would be to decouple economic growth from the consumption of natural resources, there are few countries that have managed to partially solve this problem (France, Hungary, Japan, Slovakia, Spain) (van der Voet *et al.*, 2005). The amount of waste generated and its composition are not constant, but depend on the structure of the economy, the level of development-innovation, investments in science, research, clean technologies (OECD, 2020). To support cities and regions in waste management, the *Interreg Europe Environment and Resource Efficiency Policy Learning Platform* has published a policy brief on sustainable waste management in a circular economy (Interreg Europe, 2020).

The evolution from a linear to a circular economy is a major challenge that requires a much stronger commitment to sustainable waste and resource management. First, less waste needs to be generated through the development of sustainable production and products and the massive implementation of an infrastructure for reuse and repair. Secondly, investment is needed in higher recycling capacities, especially for food waste, plastic and packaging. Finally, incineration and landfilling must be kept to a minimum, while solving the problem of European landfills in order to regain land and ecosystems (OECD, 2020).

3.2. Transforming waste into resources in the context of circular economy model

At European level, a number of countries are developing on the basis of a linear economy, where products are generated, used and disposed of or incinerated, often after a single use. Today, only 12% of the materials used by European industry come from recycling (Waltersson, 2022). Under the European Green Deal, the European Commission points out that from 1970 to 2017, the annual global extraction of materials has tripled and continues to increase, posing a major risk to future generations. About half of greenhouse gas emissions and more than 90% of biodiversity loss and water stress come from resource extraction and food and food processing (COM 640, 2019).

Indeed, humanity now uses the nature of about 1.9 times faster than our planet's ecosystems can regenerate. European Commission adopted in December 2015 the *Circular Economy Action Plan*, to advance employment, development and investment and to ensure a competitive and resource-efficient economy (European Union, 2020). On 23 February 2018, the European Union approved the four legislative proposals of the waste package (Waste Framework Directive, Packaging Waste Directive, Landfill Directive, New Harmonized EU Methodology for Calculating Municipal Waste Recycling Rates), which aspires to lead to greater waste recycling and contribute to the creation of a circular economy.

The agreements set binding waste reduction targets and updated rules to reduce waste production, ensure better control of waste management, encourage product reuse and improve recycling in all EU countries. On 4 March 2019, it was published a comprehensive report highlighting that 54 actions under the action plan have been completed or are being implemented, including new European waste legislation, with new ambitious recycling targets (OECD, 2020). On 11 March 2020, the European Commission assumed a new action plan for circular economy including new initiatives along the product life cycle that should ensure a foremost modernization and transformation of the European economy, together with environmental protection and giving new rights to consumers. The plan is one of the main elements of the European Green Deal which sets out an ambitious roadmap to a climate-neutral circular economy (COM 68, 2020).

4. Recovery of waste materials - as secondary sources

4.1. The concept of "urban mining" and its implications

The daily reality has shown that, as the demand for raw materials increases, more efforts will have to be made for recycling. Higher recycling rates will ensure the following advantages: reduce the pressure on demand for primary

raw materials; helps to reuse valuable materials that would otherwise be wasted; reduce energy consumption, greenhouse gas emissions and other negative impacts on the extraction and processing environment.

To successfully recover raw materials from urban mines, metal recycling entails a systematic recycling approach with efficient recycling schemes, together with a deep knowledge of the complexity of the interactions between different materials in waste streams. "Urban mining" is an exciting equivalence for the development of large-scale recycling of urban waste. In early 1969, Jane Jacobs launched this idea in her book *The Economy of Cities* where she considered that "cities are the mines of mineral reserves." In 1988, the Japanese professor Randolph Nanjo created the term "urban mine" to identify selected places where waste of electric and electronic equipment as well as mechanical equipment can be deposited and where metals can be extracted from these products considered to be at the end of life (Qi *et al.*, 2016). The model has been extensively accepted in Japan. In the 1980s, Chinese researchers proposed the concepts of "urban minerals" and "municipal minerals" with an emphasis on the recovery and reuse of scrap metals (Zhang *et al.*, 2012).

Urban mining involves the recovery of valuable components as raw materials from secondary resources (<https://www.sintef.no/en/urban-mining/>). "*Urban minerals address recyclable resources such as steel, non-ferrous metals, rare metals, plastics and rubber contained in waste mechanical and electrical equipment, wire and cable, communication tools, automobiles, household appliances, electrical products, metal packages and plastic and other waste materials that have been generated in the industrialization and urbanization processes. The amounts accessible are almost comparable to those of primary ore resources. Urban minerals are generated in cities as solid waste that is used and reused as recyclable resources*" (Qi *et al.*, 2016).

The permanent demand for energy, materials and services has resulted in substantial accumulations of natural resources in buildings, infrastructure, products and landfills (Brunner and Rechberger, 2004; UNEP, 2020). It has been found at present that, for example, the copper or iron reserves remaining in geological ores is equivalent to that of potential metallic waste and products (buildings, infrastructure, products, different waste, landfills) (Kapur and Graedel, 2006; Müller *et al.*, 2006; Spatari *et al.*, 2006; UNEP, 2020). These resources were beforehand used, but they are not lost, being found in landfills, slag piles, tail ponds, WEEE etc. (Brunner, 2011; Ghiga *et al.*, 2020; Krook and Baas, 2013; Tătaru-Fărnuș *et al.*, 2019).

Attention is currently being paid to the various aspects of two such emerging approaches, which go beyond the end-of-life phase: landfills (mining in landfills) and recycling of annual waste streams (urban mining). Although the resource potential of stocks of metals in use in terms of future waste streams is often specifically emphasized, references are more or less made to the recovery

of metals (but also other resources) from anthropogenic sources (Ayres *et al.*, 2001; Brunner and Rechberger, 2004; Klinglmair and Fellner, 2010; Wittmer and Lichtensteiger, 2007).

Urban mining is largely similar to other concepts already used, such as resource management, closing loops, the cradle-to-cradle principle and integrated waste management, eco-innovation and eco-design (Baccini and Brunner, 2012). In principle, waste mining refers to the processing, treatment and recovery of stored materials, informally located in landfills and structured landfills (Savage *et al.*, 1993).

4.2. Circular economy, secondary raw materials in the EU and critical raw materials

Recycling can provide many secondary raw materials (SRM) and facilitate the transition to a more circular economy at EU level and meet the demand for raw materials in the EU (Mathieux *et al.*, 2017). Although all raw materials are imperious, some of them are of greater interest than others in terms of safe and sustainable supply. These are denoted as **critical raw materials (CRM)**.

The list of CRM for the EU and the basic methodology of the European Commission (EC) criticality assessment become key tools in the context of EU raw materials policy. CRMs are of great economic importance to the EU and present a high risk of supply disruption. Raw materials are essential for the production of a wide range of goods and applications used in everyday life. They are intrinsically linked to all industries at all stages of the supply chain. They are crucial for a strong European industrial base, a key element of EU growth and competitiveness. Accelerating cycles of technological innovation and the rapid growth of emerging economies have led to a steadily growing demand for these highly sought after metals and minerals. Future use of global resources could double between 2010 and 2030.

At present, Europe faces a major risk of providing critical raw materials, which can generate major supply chain problems and significant impacts on the economy compared to other raw materials (Hennebel *et al.*, 2015). Within the EU Innovation Partnership on Raw Materials Initiative, European Commission has developed a list of critical materials (Fig. 3), which includes elemental metals (such as those from the platinum group and rare earth elements), but also industrial minerals (Novák *et al.*, 2021). To overcome these risks, an innovative alternative to conventional mining was advanced, which would ensure the need for critical raw materials through urban mining. In this context, to address the growing concern about securing valuable raw materials for the EU economy, the Commission launched the European Raw Materials Initiative in 2008 as an integrated strategy setting out targeted measures to ensure and improve access to raw materials for the EU, as a result of the fact that CRMs are absolutely

necessary for high-tech products, emerging innovations, technological progress and increasing quality of life.

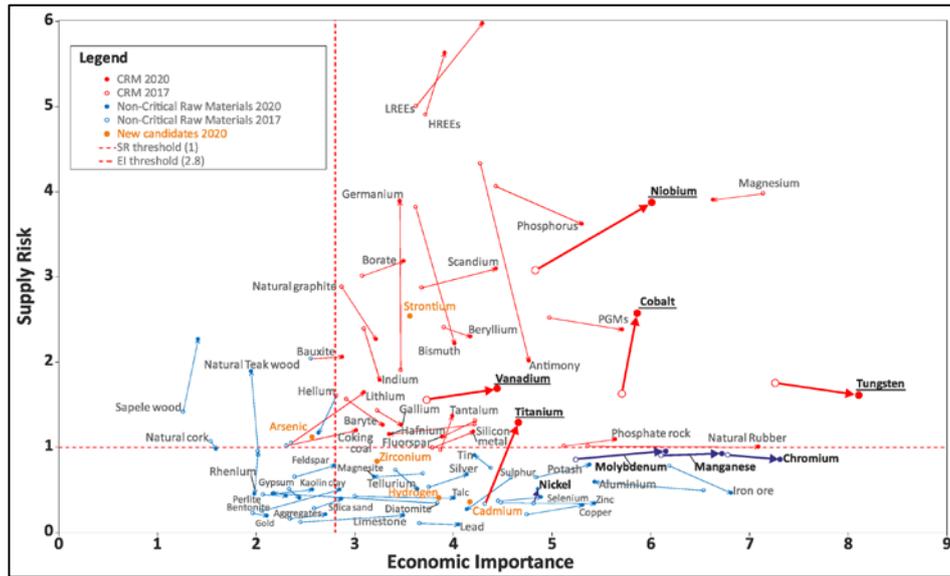


Fig. 3 – Assessment of the criticality dynamics in 2017–2020 (individual materials and groups) (Novák *et al.*, 2021; reproduced under the terms and conditions of the Creative Commons Attribution (CC BY) license <http://creativecommons.org/licenses/by/4.0/>).

A relevant example is given by Gislev *et al.* (2018) who considered a smartphone which can contain a large number of different metals with various properties (up to 50), with light weight and small dimensions. CRMs are absolutely necessary for solar panel components, electric cars, wind turbines, which are considered environmentally friendly and for climate change mitigation. Moreover, the production of low-carbon technologies in EU necessary to meet the climate and energy targets can increase significantly (by a factor of 20) the request for certain raw materials by 2030 (Hafner and Raimondi, 2020). Although many CRMs have a high recycling potential and, despite government encouragement to move to a circular economy, the end-of-life rate of re-entry of CRMs is generally low (Gislev *et al.*, 2018) (Fig. 4).

There is CRM, such as antimony, cobalt, vanadium, tungsten, etc. with a high recycling rate, due to the fact that the collection rate is high mainly because there is legislation in this regard, and the demand is also very high in the machine tool industry, batteries, industrial catalysts, automobiles. But globally, addressing all CRMs, the recycling rate is low, being around 14% for platinum metals.

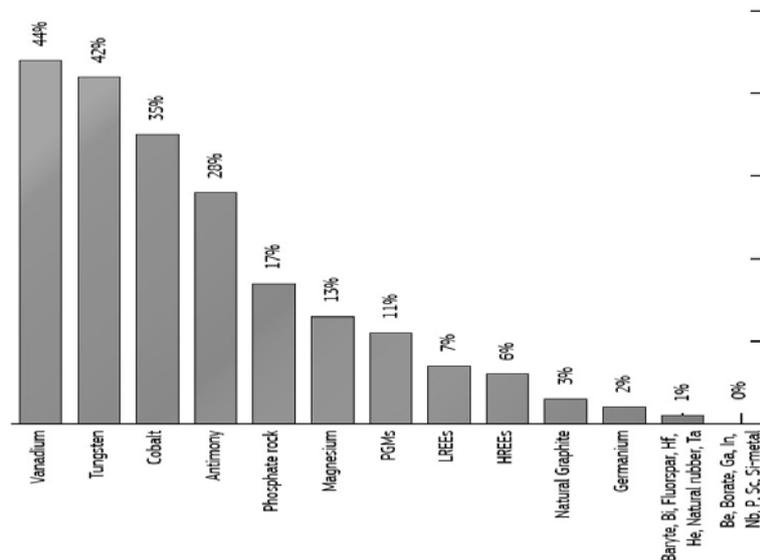


Fig. 4 – Current contribution of recycling to meet EU demand of CRMs: End-Of-Life recycling Input Rate (adapted upon Gislev *et al.*, 2018).

European industry is characterized by a series of imbalances, both in the extraction-processing stages (upstream of the production process itself) and in the use processes (downstream), due to the fact that the value chain of CRM cannot be fully insured from conventional sources, these being located only in a few areas, some characterized by regional or political instability (Fig. 5). These imbalances sharply pose the problem of alternative sources to meet the vast need for CRM so as to ensure both the continuation of production processes in European industrial systems as well as social security (Gislev *et al.*, 2018).

Just as the extraction of primary CRMs in Europe helps to ensure the security of supply of raw materials to European industry, so is the case for the efficient management of secondary resources throughout the life cycle and the recycling of waste in secondary CRMs. Consequently, substitution and recycling are considered risk reduction measures in the methodology for drawing up the EU list of critical raw materials (EC, 2016; EC, 2018).

To date, researchers have not fully realized that a major potential of resources is in urban stock, municipal solid waste landfills (MSW) and other waste, or in tailings and mining waste, or polluted soils with metals (Minut *et al.*, 2021). Finally, economic modeling is needed to clarify which potentials of secondary resources promise an economic benefit and which of them are marginal or even negative assets.

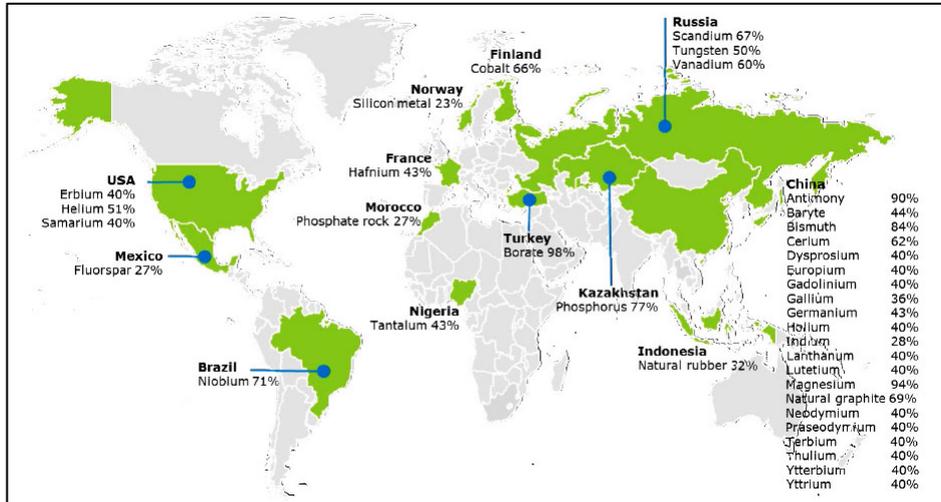


Fig. 5 - Contribution of primary global suppliers of critical raw materials, average from 2010-2014 (Gislev *et al.*, 2018).

5. Methods for the recovery of secondary and critical raw materials from waste

The recovery of CRM from waste and residues of the global mineral industry, as secondary sources, offers the possibility to establish stable international supply routes to cover the needs of existing technologies with critical raw materials and allow the development of future advanced applications, which will significantly reduce the impact on the environment.

5.1. Pyrometallurgy

Pyrometallurgy is actually a branch of extractive metallurgy, consisting of thermal treatment of minerals and ores and metallurgical concentrates (calcination, frying, melting, refining) to produce physical and chemical transformations into materials that allow the recovery of valuable metals. Pyrometallurgical treatment (Fig. 6) can generate products that can be sold (pure metals as iron, copper, zinc, chromium, tin and manganese), intermediate compounds or alloys, suitable for further processing. Unfortunately, pyrometallurgical processes require high energy consumptions, usually supplied by combustion or using electricity (Rai *et al.*, 2021).

In addition to metals and metal compounds in the process, slag, flue gases and soot also result. The flue gas is composed of dust, smoke and toxic gases at high temperatures. These harmful substances are transformed into useful products through the complete pyrometallurgical use of flue gases and soot.

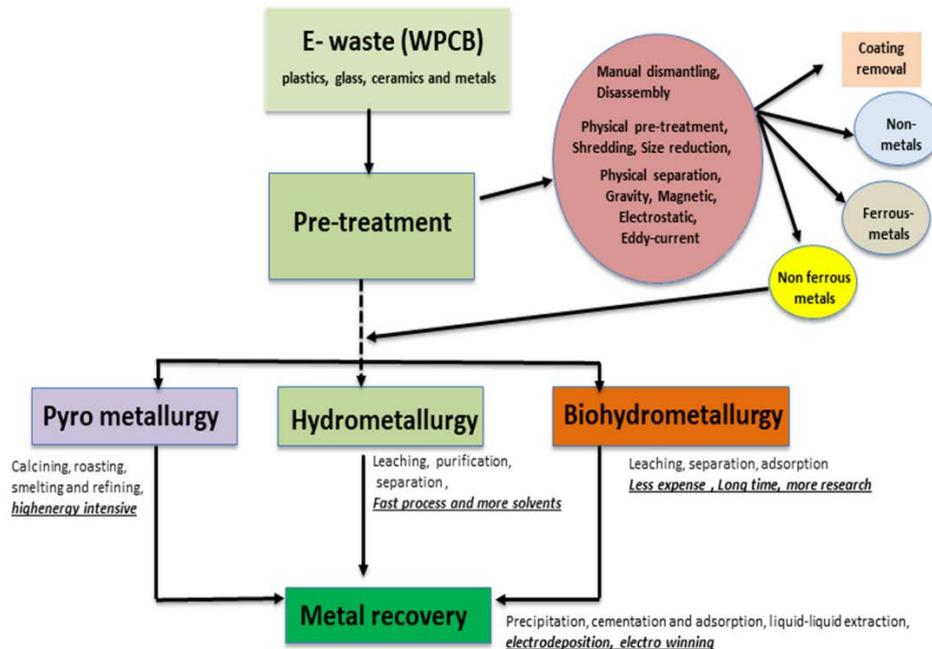


Fig. 6 – Scheme of metal recovery from e-waste (printed circuit boards) (Rai *et al.*, 2021; reproduced under the terms and conditions of the Creative Commons Attribution (CC BY) license <http://creativecommons.org/licenses/by/4.0/>).

5.2. Hydrometallurgy

Hydrometallurgy is a method related to extractive metallurgy, for obtaining metals from their ores in three steps: separation by leaching; separation by concentration and purification of the solution; recovery of metals or metal compounds. The hydrometallurgical process originates from the 16th century, but it has been developed only in the 20th century, partially stimulated by gold extraction from ores with a low degree of purity. An extremely wide range of applications of hydrometallurgy has been reached, as scientific technology has advanced through the development of ion exchange processes, solvent extraction and other processes. Now hydrometallurgy is applied for the production of over 70 metallic elements (gold, silver, copper, zinc etc.) (Apostolescu *et al.*, 2022; Sethurajan *et al.*, 2019). Hydrometallurgy is also applied for the recovery of CRM from WEEE within at least two main operations, namely (1) leaching (solubilization of metals from WEEE into leachate using aqueous chemicals) and (2) recovery (selective recovery of dissolved metals from leachate) (Fig. 6).

5.3. Biotechnological processes applied in the extraction, processing and recycling of critical raw materials

Biotechnology can significantly contribute to the recovery of CRM from secondary resources, being able to ensure selective, efficient, scalable processes that allow the procurement of products with a very good degree of purity or to favor the continuous modernization of products. These processes are based on biomining by using autotrophic and heterotrophic microorganisms and applying extracellular precipitation of metals with biogenic compounds, bioelectrochemical recovery of metals, intracellular biosynthesis of solid metals, microbial and methylation sorption (Hennebel *et al.*, 2015). Microorganisms are applied in biohydrometallurgy (Fig. 6), when biomining is based on the oxidation of minerals with reduced sulfides by acidophilic chemo-autolithotrophic microorganisms, or biooxidation, or bioleaching (release of the desired element from the ore), or reductive dissolution of oxide ores.

Biohydrometallurgy processes are a cost-effective and environmentally friendly alternative to conventional processes (hydrometallurgy) (Auerbach *et al.*, 2019; Gavrilesu, 2022). Bioleaching is the biological conversion of insoluble metal compounds into a water-soluble form using microorganisms (Sand *et al.*, 2001) and is based on the mobilization of metal ions by oxidation and biological complexation (Rohwerder *et al.*, 2008). The microorganisms involved have several functions in this process. For example, when recovering metals from magnets, firstly, they generate ions and/or protons, Fe(III) and secondly concentrate them at the interface between the material and the bacterial cell. Thus, it leads to an increase in the leaching of the sample material. Microorganisms with different types of metabolism can be used: on the one hand microorganisms that produce inorganic or organic acids to dissolve the material directly, and on the other hand microorganisms that use magnet iron as an electron donor. This leads to an oxidation of the iron and therefore to a destruction of the matrix, from where other metals could enter the solution.

Due to increasing demand for raw materials and rising prices, interest in bio-leaching of secondary raw materials has increased. Various secondary sources for the recovery of critical metals through bioleaching have already been studied and are moving towards an industrial implementation.

6. Case study: recovery of cobalt from secondary sources

6.1. Current situation of Cobalt resources in Europe

Cobalt is the 29th most abundant element in the earth's crust, with a concentration of about 25 ppm. Usually, cobalt is obtained as a by-product in the processing of ores of other metals, in particular copper and nickel, which account for 35% and 55% of production, respectively. The remaining 10% of world

production comes from primary cobalt operations (EC, 2018). The world resources of Co are estimated at about 15 million tons. The most important resources are in the Copper Belt of Central Africa, Congo and Zambia, estimated at over 6 million tonnes of cobalt. An additional 1 billion tons of cobalt resources can be found in manganese crusts on the ocean floor, which are not economically exploitable (Petavratzi *et al.*, 2019; USGS, 2013). In Europe there are only smaller known cobalt resources (for example, in the Kupferschiefer), but there is currently no significant mining of this resource.

The European Union is currently an importer of ore and cobalt concentrates (Fig. 7), but there has been a decrease in imports in recent years as a result of policies to recover it from secondary resources (Petavratzi *et al.*, 2019).

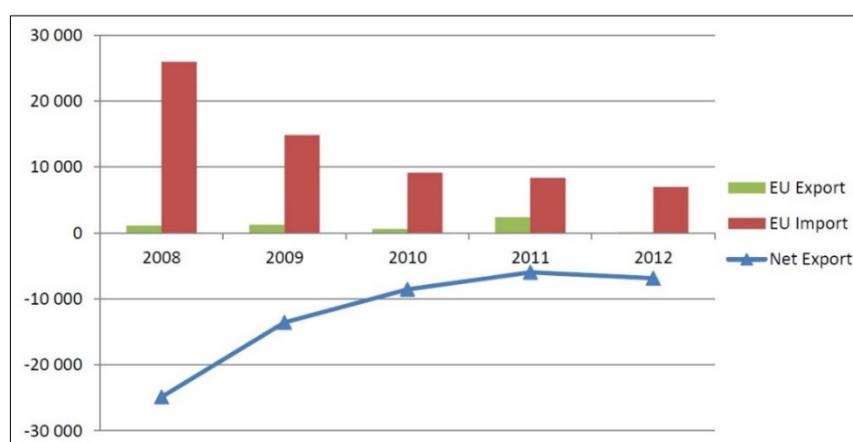


Fig. 7 – Trends in extra-EU trade in cobalt ores and concentrates (tonnes) (Eurostat-Comext Database).

The supply chain for the European cobalt industry is shown in Fig. 8. As can be seen, the EU imports ores and concentrates, impure carbonate and impure hydroxide residues, as well as non-EU metal to produce cobalt powder and cobalt compounds. Applications in the field of cobalt-related biotechnology are becoming more numerous, as it is an essential biological nutrient for fermentation processes that are used in several sectors.

6.2. Recovery of cobalt from waste lithium batteries

Batteries are an important secondary source of metals and therefore the recycling of lithium (LiB) batteries ensures both environmental protection, but saves natural resources and helps in reducing battery manufacturing costs, especially as the concentration of metals in waste batteries is often superior to that of natural ores. Cobalt is a valuable metal that can also be recovered from

batteries, which is a major advantage, as much of the global demand for cobalt is for batteries (25% in 2017), and the price of cobalt has risen significantly in recent years (Dewulf *et al.*, 2010; Dorella and Mansur, 2007; Jha *et al.*, 2013).

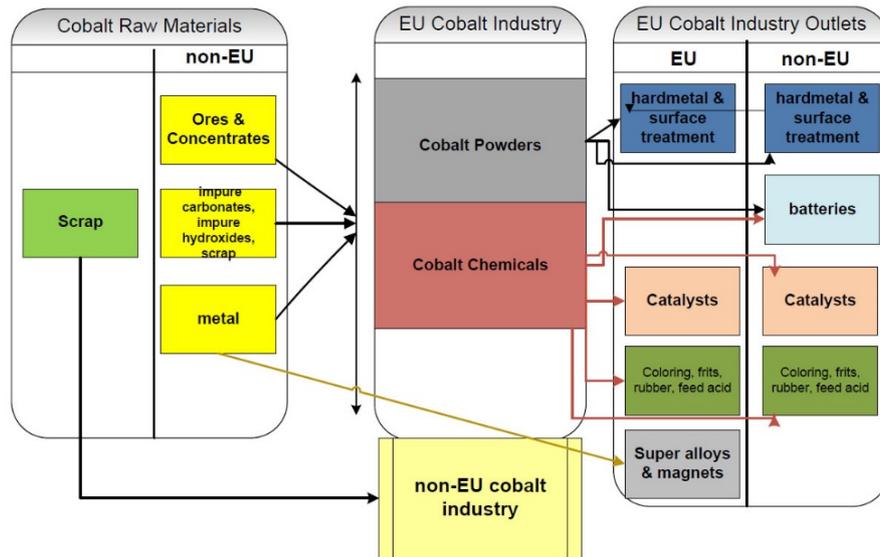


Fig. 8 – The supply chain for the European cobalt industry (EC, 2015).

LiB batteries are widely used because they have a number of advantages compared, for example, with nickel-cadmium batteries: they can provide high energy densities, have a relatively low weight, low self-discharge rate, an operating temperature range relatively wide etc. The typical composition of these batteries consists in: 5-20% by weight Co, 5-10% by weight Ni, 5-7% by weight Li, 15% by weight organic substances and 7% by weight plastics. LiCoO_2 powder is widely used as a cathode in LiB, due to its good electrochemical performance, but its use is limited by the limited resources of cobalt and its toxicity (Bertuol *et al.*, 2016; Kang *et al.*, 2010; Li *et al.*, 2013).

Two different classes of processes can be distinguished depending on the secondary source used for the recovery of cobalt from used LiB: **pyrometallurgical processes and hydrometallurgical processes.**

Pyrometallurgical processes allow the recovery of very valuable metals (Co, Ni in the case of LiB batteries). These processes involve the direct melting of battery waste in plants that are not necessarily designed for LiB recycling (*e.g.* metallurgical plants), which is a major industrial advantage (Georgi-Maschler *et al.*, 2012). Unfortunately, metallurgical processes induce negative impacts on the environment due to emissions of pollutants (gases, particulate pollutants) and consume a lot of energy (Jha *et al.*, 2013). They also do not allow the recovery of

low-value metals and non-metallic materials (plastics and electrolytes) (Pagnanelli *et al.*, 2016)

Hydrometallurgical processes involve the extraction of metals by leaching electrode powder, followed by refining and recovering metals. Unlike pyrometallurgy, hydrometallurgy makes it possible to recover every component present inside the batteries (both metals and non-metals). The quality and yield of the obtained materials depends on the mechanical parameters of pretreatment (for example, the type of crushing) (Jha *et al.*, 2013).

Biotechnology can contribute to the immunization of the hydrometallurgical process through **bio-hydrometallurgy** which confers a number of advantages to CRM recovery, such as higher efficiency, lower costs. The disadvantage of bio-hydrometallurgy refers to the fact that the treatment period is long and the necessary microbes are difficult to incubate efficiently (Jha *et al.*, 2013; Li *et al.*, 2013). Some studies have shown that a culture of *A. ferrooxidans* can produce sulfuric acid to leach metals indirectly from LiB Mishra *et al.* (2008). The purity of lithium and cobalt obtained in these processes was high, but the treatment period was very long and the treatment of bacteria was difficult.

Some recovery processes combine pyro- and hydrometallurgical steps and often have integrated pretreatment steps, such as pyrolysis or mechanical processing, i.e. crushing and separation of materials (Georgi-Maschler *et al.*, 2012; Velázquez-Martínez *et al.*, 2019a). Through these combinations several metals can be separated (Fig. 9). For example, Accurec GmbH® (Krefeld, Germany) applied a combination of mechanical, pyrometallurgical and hydrometallurgical processes for the recovery of a Li_2CO_3 cathode precursor and a Co-Ni-Mn alloy (Fig. 10).

The process involves: sorting used batteries; transport of disassembled material; heat treatment at 250°C and vacuum for the removal of electrolytes, solvents and volatile hydrocarbons; grinding; mechanical separation on site, magnetic separators, classifiers when resulting fractions of Fe - Ni, Al and Al - Cu, from which the base metals can be extracted; the remaining fraction is sent to agglomeration in a two-stage pyrometallurgical process, in a rotary kiln at 800°C, followed by an electric arc furnace. According to Georgi-Maschler *et al.* (2012), the Co alloy recovered at this stage has commercial value, but Mn and Li are lost in the slag or volatilized in the electric arc furnace. However, the slag contains a concentration of Co of five times higher than in the material introduced for processing, so there is a concentration of the metal, which is why the slag is further processed to obtain the cathodic precursor Li_2CO_3 , recovered in proportion of 90% (Velázquez-Martínez *et al.*, 2019b).

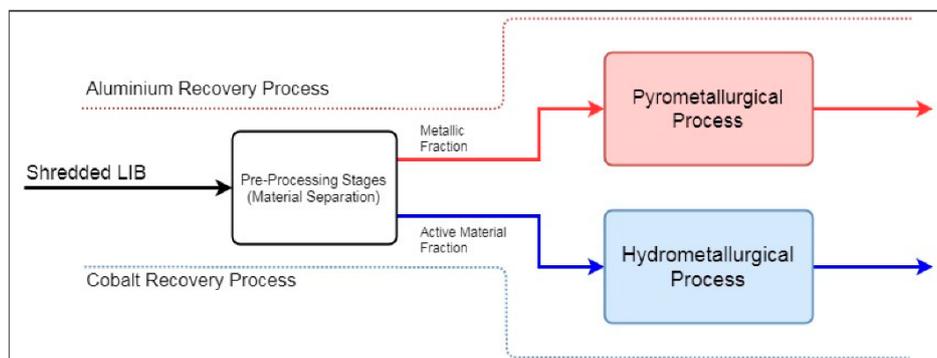


Fig. 9 – Combined scheme of pyrometallurgy and hydrometallurgy processes for the recovery of used lithium-ion batteries (Velázquez-Martínez *et al.*, 2019a; reproduced under the terms and conditions of the Creative Commons Attribution (CC BY) license <http://creativecommons.org/licenses/by/4.0/>).

Mechanical separation produces fractions of Fe - Ni, Al and Al - Cu, from which base metals can be extracted, while the remaining fraction is sent to agglomeration and to a two-stage pyrometallurgical process, first in a rotary kiln, at 800°C, and then in an electric arc furnace (EAF), where graphite is consumed to improve the recovery of Co or Mn (Georgi-Maschler *et al.*, 2012). The Co alloy recovered at this stage has commercial value. Mn and Li lost in slag or can be recovered by hydrometallurgical means in the form of the cathodic precursor Li_2CO_3 , since the concentration of Li in was estimated to be 5 times higher than in the material introduced in the process (Velázquez-Martínez *et al.*, 2019b). The Accurec process claims to have achieved a recovery of 90% Li_2CO_3 ; it can be used either as a cathodic precursor or as a raw material for the manufacture of glass. A process proposed by Bertuol *et al.* (2016) on a laboratory scale use the extraction of cobalt with supercritical fluids, to improve the leaching process, shorten the extraction time and reduce the consumption of reagents.

All tests, performed in batch mode, in a laboratory installation were made under CO_2 pressure of 75 bar, at a temperature of 75°C. The results showed that the extraction of cobalt with supercritical liquids from used lithium-ion batteries offers advantages over conventional methods. Supercritical extraction allowed the recovery of 95.5% by weight of cobalt in a shorter reaction time and the use of a smaller amount of chemical agents, compared to leaching at atmospheric pressure (Bertuol *et al.*, 2016).

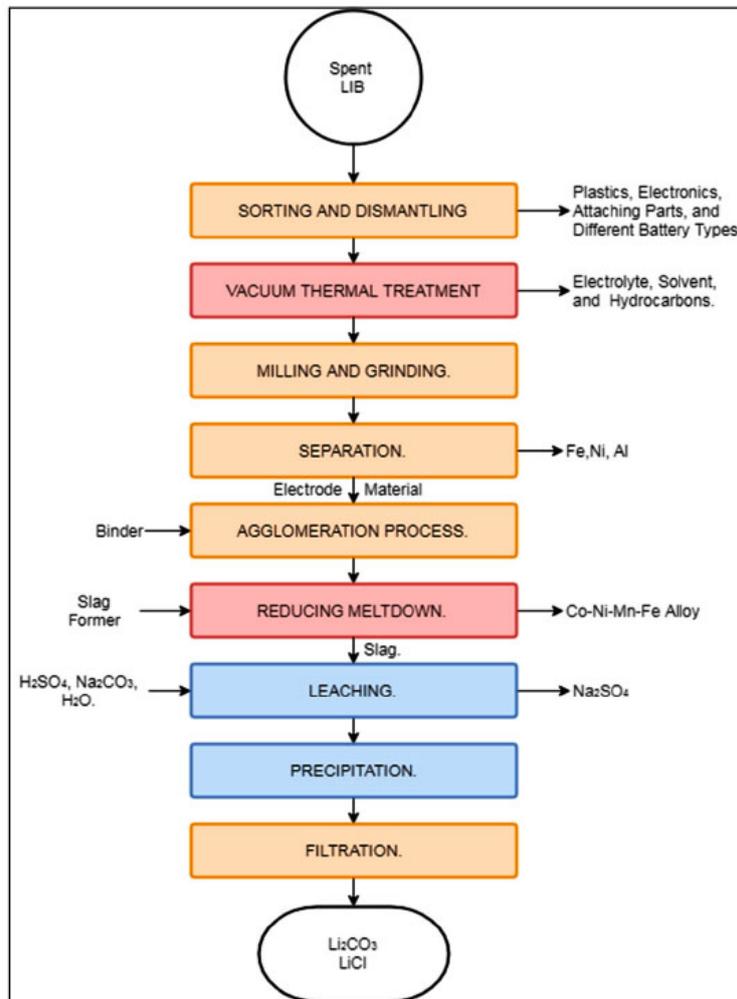


Fig. 10 – Schematic representation of the Accurec process (Velasquez *et al.*, 2019b; reproduced under the terms and conditions of the Creative Commons Attribution (CC BY) license <http://creativecommons.org/licenses/by/4.0/>).

7. Conclusions

An analysis of European municipal waste management practices has shown that municipal waste is a largely untapped source of recyclable materials for production, reusable goods, as well as a source of heat and electricity when used properly in efficient plants which convert waste in energy. Along with the many environmental and health problems caused by current patterns of consumption and waste disposal, they must be addressed in such a way as to

become a secondary source of raw materials, in order to contribute to the development of viable and sustainable cities of tomorrow.

The waste management framework established by the Waste Framework Directive places priorities for waste management with the preferred waste prevention option, followed by reuse and recycling, which closes the product life cycle. At the bottom of the hierarchy is the recovery of energy from waste, the option of storing landfills is the least desired. Therefore, in order to improve waste management, actions are a priority according to the "waste hierarchy".

Europe is facing an increasing risk of supply of critical raw materials. They can be defined as materials whose supply chain risks and their impact on the economy are higher compared to most other raw materials, which is why they have been designated as critical raw materials. In order to meet the challenge of supply risk, an innovative approach to sustainable primary mining, substitution of critical metals and urban mining was considered necessary at European level.

Landfill mining is proposed as an innovative strategy for mitigating the environmental risks associated with landfills, but also for recovering raw materials and secondary energy from landfilled waste. Landfills have been and are used for a variety of wastes, currently representing an accumulation of large quantities of very different materials, including critical raw materials (CRM) and other valuable materials. Therefore, the recovery of secondary raw materials from landfills can help reduce the risks posed by the lack of critical raw materials.

Data on trade flows in recent years show that the EU has been a net importer of cobalt ores and concentrates due to lack of production within the EU. Cobalt is recycled for economic reasons (lower costs compared to cobalt extraction from ores) and for environmental reasons (to prevent its damage caused by eg the presence of used batteries). Two different classes of processes can be distinguished depending on the secondary source used for the recovery of cobalt from used LiB: pyrometallurgical processes and hydrometallurgical processes. The bio-hydrometallurgical process has some advantages over the hydrometallurgical process, such as higher efficiency, lower costs and some industrial requirements, but the treatment period is long and the necessary microbes are difficult to incubate efficiently.

Some recovery processes combine pyro- and hydrometallurgical steps and often have integrated pretreatment steps. Laboratory, pilot, and industrial scale processes have shown that LiBs are important sources of lithium, cobalt, and a few other critical materials.

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REFERENCES

- Apostolescu N., Vizitiu M.A., Cernătescu C., Tătaru-Fărnuș R.E., Cobzaru C., Apostolescu G.A., *Current trends in the recovery of precious metals from electrical and electronic waste*, Buletinul Institutului Politehnic din Iasi – Sectia Chimie și Inginerie Chimică, **68(72)**, 51-74 (2022).
- Ayres R.U., van den BerghJeroen J.C.J.M., van den BerghJohn C.J.M., Gowdy M., Gowdy J.M., *Strong versus weak sustainability: Economics, natural sciences, and "consilience"*, Environmental Ethics, **23**, 155-168 (2001).
- Anghelescu A., Ponoran I., Ciubotaru V., *Mediul ambiant si dezvoltarea durabila (Environment and Sustainable Development)*, ASE Publishing House, Bucharest, Romania, 2003.
- Auerbach R., Bokelmann K., Stauber R., Gutfleisch O., Schnell S., Ratering S., *Critical raw materials - Advanced recycling technologies and processes: Recycling of rare earth metals out of end of life magnets by bioleaching with various bacteria as an example of an intelligent recycling strategy*, Minerals Engineering, **134**, 104-117 (2019).
- Baccini P., Brunner P.H., *Metabolism of the Anthroposphere: Analysis, Evaluation, Design* (2nd edition), The MIT Press, 2012.
- Bertuol D.A., Machado C.M., Silva M.L., Calgaro C.O., Dotto G.L., Tanabe E.H., *Recovery of cobalt from spent lithium-ion batteries using supercritical carbon dioxide extraction*, Waste Management, **51**, 245-251 (2016).
- Brunner P.H., Rechberger H., *Practical Handbook of Material Flow Analysis*, Lewis Publishers, Boca Raton, FL, USA, 2004.
- Brunner P.H., *Urban mining. A contribution to reindustrializing the city*, Journal of Industrial Ecology, **15**, 339-341 (2011).
- COM 640, *Communication from the Commission to the European Parliament, the European Council, the Council, the European Economic and Social Committee and the Committee of the Regions. The European Green Deal*, European Commission, Brussels (2019).
- Dewulf J., Van der Vorst G., Denturck K., Langenhove H.V., Ghyoot W., Tytgat J., Vandeputte K., *Recycling rechargeable lithium ion batteries: critical analysis of natural resource savings*, Resource Conservation and Recycling, **54**, 229–234, (2010).
- Dorella G., Mansur M.B., *A study of the separation of cobalt from spent Li-ion battery residues*, Journal of Power Sources, **170**, 210–215 (2007).
- EC, *Being Wise with Waste: The EU's Approach to Waste Management*, European Commission, Brussels, Belgium, (2010).
- EC, Report on Critical Raw Materials for the EU Critical Raw Materials Profiles, <https://ec.europa.eu/docsroom/documents/11911/attachments/1/translations> (2015).
- EC, *Raw Materials Scoreboard. European Innovation Partnership on Raw Materials*, European Commission, Luxembourg (2016).
- EC, *Report on Critical Raw Materials and the Circular Economy*, European Commission, Brussels, On line at: http://publications.europa.eu/resource/cellar/d1be1b43-e18f-11e8-b690-01aa75ed71a1.0001.01/DOC_1 (2018).

- EC Directive 98, *Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives* (Text with EEA relevance), Official Journal of the European Union, L 312/3 (2008).
- EEA, *Resource Efficiency and Waste*, European Environment Agency, Copenhagen, Denmark (2019).
- European Union, *Circular Economy Action Plan. For a Cleaner and More Competitive Europe*, European Commission, Luxembourg (2020).
- Eurostat, *Municipal waste statistics*, On line: https://ec.europa.eu/eurostat/statistics-explained/index.php/Municipal_waste_statistics (2019).
- Everett J.W., *Solid Waste Disposal and Recycling, Environmental Impacts*, In: *Encyclopedia of Sustainability Science and Technology*, Meyers R. (Ed.), Springer, Cham, Switzerland (2012).
- Gavrilescu M., *Microbial recovery of critical metals from secondary sources*, *Bioresource Technology*, **344**, 126208, <https://doi.org/10.1016/j.biortech.2021.126208> (2022).
- Georgi-Maschler T., Friedrich B., Weyhe R., Heegn H., Rutz M., *Development of a recycling process for Li-ion batteries*, *Journal of Power Sources*, **207**, 173-182 (2012).
- Ghiga S.C., David M., Minuț M., Comanita-Ungureanu E.D., Cozma P., Gavrilescu M., *Reducing Health and Ecological Risks by Using WEEE as Secondary Sources for Critical Raw Materials*, IEEE 2020 International Conference on E-Health and Bioengineering (EHB), Book Series, E-Health and Bioengineering Conference, October 29-30, <http://doi.org/10.1109/EHB50910.2020.9280253>, (2020).
- Ghinea C., Cozma P., Gavrilescu M., *Artificial neural network applied in forecasting the composition of municipal solid waste in Iasi, Romania*, *Journal of Environmental Engineering and Landscape Management*, **29**, <https://doi.org/10.3846/jeelm.2021.15553> (2021).
- Gislev M., Grohol M., et al., *Report on Critical Raw Materials and the Circular Economy*, European Commission, Directorate-General for Internal Market, Industry, Entrepreneurship and SMEs, Brussels (2018).
- Hafner M., Raimondi P.P., *Priorities and challenges of the EU energy transition: From the European Green Package to the new Green Deal*, *Russian Journal of Economics*, **6**, 374-389 (2020).
- Hennebel T., Boon N., Maes S., Lenz M., *Biotechnologies for critical raw material recovery from primary and secondary sources: R&D priorities and future perspectives*, *New Biotechnology*, **32**, 121-127 (2015).
- Interreg Europe, *Sustainable Waste Management in a Circular Economy. A Policy Brief from the Policy Learning Platform on Environment and Resource Efficiency*, European Regional Development Fund, European Union, Brussels (2020).
- Jha A.K., Jha M.K., Kumari A., Sahu S.K., Kumar V., Pandey B.D., *Selective separation and recovery of cobalt from leach liquor of discarded Li-ion batteries using thiophosphinic extractant*, *Separation and Purification Technology*, **104**, 160-166 (2013).
- Kang J., Senanayake G., Sohn J., Shin S.M., *Recovery of cobalt sulfate from spent lithium ion batteries by reductive leaching and solvent extraction with Cyanex 272*, *Hydrometallurgy*, **100**, 168-171 (2010).

- Kapur A., Graedel T.E., *Copper mines above and below ground. Estimating the stocks of materials in ore, products, and disposal sites opens up new ways to recycle and reuse valuable resources*, *Environmental Science & Technology*, **40**, 3135-3141 (2006).
- Klinglmair M., Fellner J., *Urban mining in times of raw material shortage*, *Journal of Industrial Ecology*, **14**, 666-679 (2010).
- Krook J., Baas L., *Getting serious about mining the technosphere: a review of recent landfill mining and urban mining research*, *Journal of Cleaner Production*, **55**, 1-9 (2013).
- Li L., Dunn J.B., Zhang X.X., Gaines L., Chen R.J., Wu F., Amine K., *Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment*, *Journal of Power Sources*, **233**, 180-189, 2013.
- Mathieux F., Ardente F., Bobba S. *et al.*, *Critical Raw Materials and the Circular Economy*, Joint Research Center, Ispra, Italia (2017).
- Minut M., Cozma P., Roșca M., Vasilachi I.C., Hlihor R.M., Diaconu M., Gavrilescu M., *Recovery of Valuable Heavy Metals from Polluted Soil using Phytomining Process - A new Challenge for Earning Secondary Raw Materials and Health Risk Reduction*, IEEE 2021 International Conference on E-Health and Bioengineering (EHB 2021), 9th Edition, Book Series, E-Health and Bioengineering Conference, November 18-19, <http://doi.org/10.1109/EHB52898.2021.9657624> (2021).
- Müller D., Wang T., Duval B., Graedel T.E., *Exploring the engine of anthropogenic iron cycles*, *PNAS*, **103**, 16111-16116 (2006).
- Novák P., Bellezze T., Cabibbo M., Gamsjäger E., Wiessner M., Rajnovic D., Jaworska L., Hanus P., Shishkin A., Goel G., Goel S., *Solutions of critical raw materials issues regarding iron-based alloys*, *Materials*, **14**, 899, <https://doi.org/10.3390/ma14040899> (2021).
- OECD, *Environment at a Glance Indicators – Circular Economy, Waste and Materials*, Organization for Economic Cooperation and Development, OECD, Paris, 2020.
- Qi J., Zhao J., Li W., Peng X., Wu B., Wang H., *Development of Circular Economy in China*, Springer, Singapore (2016).
- Pagnanelli F., Moscardini E., Altimari P., Abo Atia T., Toro L., *Cobalt products from real waste fractions of end of life lithium ion batteries*, *Waste Management*, **51**, 214-221 (2016).
- Petavratzi E., Gunn G., Kresse C., *Commodity Review: Cobalt*, British Geological Survey (2019).
- Rai V., Liu D., Xia D., Jayaraman Y., Gabriel J.-C.P., *Electrochemical approaches for the recovery of metals from electronic waste: a critical review*, *Recycling*, **6**, 53, <https://doi.org/10.3390/recycling6030053> (2021).
- Rohwerder T., Gehrke T., Kinzler K., Sand W., *Bioleaching review part A: progress in bioleaching: fundamentals and mechanisms of bacterial metal sulfite oxidation*, *Applied Microbiology and Biotechnology*, **63**, 239-248 (2008).
- Sand W., Gehrke T., Jozsa P.-G., Schippers A., *(Bio) chemistry of bacterial leaching – direct vs. indirect bioleaching*, *Hydrometallurgy*, **59**, 159-175 (2001).
- Savage G.M., Golueke C.G., von Stein E.L., *Landfill mining: past and present*, *Biocycle*, **34**, 58-61 (1993).

- Sethurajan M., van Hullebusch E.D., Fontana D., Akcil A., Deveci H., Batinic B., Leal J.P., Gasche T.A., Ali M., Kuchta K., Neto I.F.F., Soares H.M.V.M., Chmielarz A., *Recent advances on hydrometallurgical recovery of critical and precious elements from end of life electronic wastes - a review*, *Critical Reviews in Environmental Science and Technology*, <https://doi.org/10.1080/10643389.2018.1540760> (2019).
- Slorach P.C., Jeswani H.K., Cuéllar-Franca R., Azapagic A., *Environmental and economic implications of recovering resources from food waste in a circular economy*, *Science of the Total Environment*, **693**, 133516, <https://doi.org/10.1016/j.scitotenv.2019.07.322> (2019).
- Spatari S., Zhang Y., Maclean H.L., *Life cycle assessment of switch grass- and corn stover-derived ethanol-fueled automobiles*, *Environmental Science and Technology*, **39**, 9750-9758 (2006).
- Tătaru-Fărnuș R.-E., Cernătescu C., Apostolescu G.A., Cobzaru C., Apostolescu N., Vizitiu M.A., Baci R.-E., *Recycling electronic waste. Study for the recovery of precious metals*, *Buletinul Institutului Politehnic din Iasi – Sectia Chimie și Inginerie Chimică*, **65(79)**, 15-25 (2019).
- UNEP, *A Year in Review, Annual Report*, International Environmental Technology Centre, United Nations Environmental Program, UNEP, Paris (2020).
- USGS, *Mineral Commodity Summaries 2013*, On line at: <https://s3-us-west-2.amazonaws.com/prd-wret/assets/palladium/production/mineral-pubs/mcs/mcs2013.pdf>, (2013).
- van der Voet E., van Oers L., Moll S., Schütz H., Bringezu S., de Bruyn S., Sevenster M., Warringa G., *Policy Review on Decoupling: Development of indicators to assess decoupling of economic development and environmental pressure in the EU-25 and AC-3 countries*, European Commission, DG Environment, (2005).
- Velázquez-Martínez O., Porvali A., van den Boogaart K.G., Santasalo-Aarnio A., Lundström M., Reuter M., Serna-Guerrero R., *On the use of statistical entropy analysis as assessment parameter for the comparison of lithium-ion battery recycling processes*, *Batteries*, **5**, 41, <http://doi.org/10.3390/batteries5020041>, (2019a).
- Velázquez-Martínez O., Valio J., Santasalo-Aarnio A., Reuter M., Serna-Guerrero R., *A critical review of lithium-ion battery recycling processes from a circular economy perspective*, *Batteries*, **5**, 68, <http://doi.org/10.3390/batteries5040068>, (2019b).
- Waltersson J., *New Survey-High Demand for Recycled Materials among European Consumers*, Stena Recycling, <https://www.stenarecycling.com/news/high-demand-for-recycled-materials-among-european-consumers/> (2022).
- Wittmer D., Lichtensteiger T., *Development of anthropogenic raw material stocks: a retrospective approach for prospective scenarios*, *Minerals and Energy - Raw Materials Report*, **22**, 62-71 (2007).
- Zhang K., Schnoor J.L., Zeng E.Y., *E-waste recycling: Where does it go from here?* *Environmental Science and Technology*, **46**, 10861-10867 (2012).

MANAGEMENTUL DURABIL, TRATAREA ȘI
RECUPERAREA DEȘEURILOR ÎN CONTEXT EUROPEAN: MINERITUL
URBAN ȘI ECONOMIA CIRCULARĂ

(Rezumat)

Lucrarea oferă o privire de ansamblu asupra alternativelor de management al deșeurilor în cadrul european al modelului economiei circulare, cu accent pe utilizarea deșeurilor ca surse secundare pentru recuperarea materiilor prime și a materialelor, în special a materiilor prime critice și subliniind importanța impactului economic și de mediu al acestei abordări. În acest context, s-au avut în vedere: analiza practicilor europene de management al deșeurilor urbane; sublinierea relevanței recuperării deșeurilor în contextul economiei circulare; valorificarea materiilor prime critice (CRM) din deșeuri specifice, ca surse secundare de materii prime. În ultima parte a lucrării au fost evidențiate o serie de metode de valorificare a materiilor prime secundare și critice din deșeuri, analizând cazul recuperării cobaltului din resurse secundare, precum deșeurile de baterii cu litiu, la fel de importante și pentru litiu și alte câteva materiale critice.