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CURRENT TRENDS IN THE RECOVERY OF PRECIOUS METALS FROM ELECTRICAL AND ELECTRONIC WASTE

BY

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Abstract. The recovery of precious metals can be efficiently done from waste. This method is considered a secondary source of raw materials, minimizing the need for exploitation and processing raw materials, while reducing energy and water consumption. Waste from electrical and electronic industry, used photographic film or waste from the jewelry manufacturing industry, fall into this category and lead to the recovery of approximately 25% of the annual requirement of gold and 20% of the annual requirement of silver. The paper presents the advantages of recycling waste containing precious metals, the difficulties of the recycling process and methods of recovering noble metals by processing electrical and electronic waste, focusing on technologies launched in recent years (bio-hydro-metallurgical processes, bio-oxidation static processes, adsorption on activated carbon or polymeric membranes, selective extraction).

Keywords: e-waste, gold and silver recovery, responsible recycling, recycling benefits.

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1. Introduction

In addition to the benefits (improving living standards and increasing life expectancy), industrial developments have also brought negative aspects as climate change, destruction or reduction of the quality of the environment by increasing pollution, and consumption / depletion of natural resources.

However, the undesirable effects of pollution can be significantly reduced by recycling, which is the process of converting waste into new materials and objects, being an alternative to "conventional" waste disposal and leading to material savings, reducing greenhouse gas emissions. Also, recycling avoids the waste of useful materials and saves raw materials. The exploitation of mineral resources to obtain useful ores is an expensive and complicated process, which fluctuates over time due to economic and technological changes. If in the 1950 – 1960s the ores containing 2 - 3 g/t Au and 1.5 - 2 g/t Cu were considered unprofitable, in the 70's the threshold dropped to 1 g/t Au and 1g/t Cu. After 2000, there were again changes in this threshold, falling, for example for Cu, to less than 0.8 g/t (Constantinescu and Anastasiu, 2015).

Electrical and electronic waste is generated by equipments powered by electricity or electromagnetic fields and by equipments used to generate, transfer and measure electromagnetic fields. These may include: televisions, computers (including screens, keyboards, mouse), refrigerators, appliances, classic or mobile phones (Vats and Singh, 2015). Among the most used electronic products, mobile phones and computers have the most spectacular technological development, which increased the amount of waste produced by used mobile phones (over 4 billion) (Singh *et al.*, 2019). In addition to increasing the quality of human life, all this has the negative effect of increasing e-waste, because the general tendency of people is to buy everything that is new in terms of electronics, usually because the novelty is closely related to a better product performance (Baldé *et al.*, 2017). The energy consumed for the recovery of precious metals from electronic waste is lower compared to the energy required to obtain the same metals through primary metallurgy processes (extraction), which makes waste processing advantageous (Vats and Singh, 2015). For example, for every million recycled mobile phones, "16,000 kg Cu, 350 kg Ag, 34 kg Au and 14 kg Pd can be recovered" (Budhraj, 2016).

On average, the global consumption of electrical and electronic devices (excluding photovoltaic panels) increases annually by over 2.5 million metric tons (Mt), and after use, they generate a large amount of e-waste (Forti *et al.*, 2020). In the upcoming years is expected to reach a much higher value for both consumption and e-waste.

2. The benefits of recycling waste containing precious metals

Waste metal recycling technologies developed nowadays allow the reuse of metals countless times, minimizing the need for exploitation and processing of raw materials, while reducing energy and water consumption (Van Yken *et al.*, 2021). The increasing of global recycling levels benefits the transition to a low-carbon, resource-efficient green economy while contributing to green jobs.

Some studies suggest that recycling metals is two to 15 times more energy efficient than obtaining it from virgin ores. Improper e-waste management results in a significant loss of rare and valuable raw materials such as gold, platinum, cobalt and rare earth elements. Currently, up to 7% of the world's gold is contained in e-waste, 100 times more gold in a ton of e-waste than in a ton of gold ore (Official Journal of the European Union, 2018).

Recycling benefits increase if complex, responsible recycling methods are used. Proper recycling aims to dramatically reduce the risks to human health and to the environmental changes, that electronic products can cause. In Europe, recycled metals are returned to home companies at a low cost. Through an employed recycling system, Japanese producers have been pushed to make their products more sustainable. As many companies have been responsible for recycling their own products, this has put the responsibility of producers who need many employees to redesign their infrastructure.

Recycling electrical and electronic waste has many advantages (Electronic waste in the EU: data and figures, 2020; Singh *et al.*, 2019), such as:

- Conservation of natural resources: recycling allows the recovery of valuable materials from old electronic devices, materials that are used to make new products. So, we save energy, we reduce pollution, we reduce greenhouse gas emissions, and we save natural resources by extracting fewer raw materials from the earth. According to studies, 98% of the components of electronic devices are recyclable.

- Environmental protection, as recycling ensures the proper handling and management of toxic chemicals (Hg, heavy metals, chloro-fluorinated organic compounds used as coolants) contained in the e-waste stream.

- New jobs are being created, usually for professional recyclers.

- Significantly reduces the negative impact of landfills on the environment (less waste = less landfills). For example, metals (Fe, Pb, Ni, Cr, Al, Zn, etc.) used in electronic devices corrode and release harmful toxins into the soil. Similarly, the plastic in these devices pollutes, leading to the destruction of ecosystems and the deterioration of the "health" of the environment. In a negative scenario, the toxic substances spread into the soil and eventually infiltrate into the water sources. But when e-waste is recycled, it is not thrown in landfills, instead its components are separated, specialists establishing whether they are reusable or not.

3. Difficulties in the process of recycling waste containing precious metals

Metal recycling is widely regarded as a successful long-term strategy, but little information is available on the extent to which recycling actually takes place. Current estimates of global end-of-life recycling rates [percentage of a scrap metal that is actually recycled], recycled content (CR) and old waste ratios (SROs) [*i.e.*, the share of old waste in the total waste stream] are not sufficiently documented. Due to the increasing use of metals over time and the extended life of metals, many CR values are low and will remain valid in the near future.

The efficiency of the recycling process varies from metal to metal, depending on the type of material or the quality of the metals for which a process is optimized and it will never reach 100% due to thermodynamic and technological limitations. Due to both relatively low efficiency in collecting and processing most discarded metal products and the inherent limitations in recycling processes and also because the raw material is often relatively abundant and inexpensive, many overall recycling rates are very low: only for eighteen metals (Al, Co, Cr, Cu, Au, Fe, Pb, Mn, Nb, Ni, Pd, Pt, Rh, Ag, Sn, Ti and Zn) is greater than 50%. So far, only for Nb, Pb and ruthenium the recycled content is over 50%.

Recycling e-waste also poses dangers to the personnel who manage the materials to be recycled. This can result in danger of slipping or crushing, the release of hazardous substances, cuts, noise and toxic chemicals. For the safety of employees in e-waste recycling facilities, work protection sheets are provided for which they are aware and it is necessary to abide by them.

E-waste recycling can be attributed as part of the "formal" or "informal" economic sector. Formal recycling of e-waste involves the construction of special facilities, suitable equipment that allows the safe extraction of the necessary materials, while informal recycling of e-waste is unstructured, unregistered and illegal due to lack of proper installation and processes.

"The universe of metal recycling" is much more complex than it is said. It is also evolving rapidly and includes a number of sustainable development challenges, including:

- Coverage of product and material flows;
- Recovering the resources of post-consumer materials, including those of rare metals, contributing to the conservation of resources, security of supply and stability of metal prices.
- Adaptation to the manufacture of new products containing increasingly complex mixtures of materials, including composites.
- Protect the environment and human health from hazardous emissions caused by improper waste management or recycling practices.

4. Types of Waste Containing Precious Metals

Electrical and electronic waste

Electrical and electronic waste is an emerging issue for both developed and developing countries. As regulations have not yet been developed for the recycling of this waste or an appropriate collection system for recycling, the issue is quite current. This waste is dangerous for the environment but from another point of view it is valuable for recovering metals.

The precious metal scrap metal recovery market is estimated at \$ 9.4 billion by 2020 and is projected to reach \$ 11.8 billion by 2025, at an annual growth rate of 4.6%, from 2020 to 2025 (EU e-waste: data and figures (infographic), 2020).

Personal devices are the biggest contributors to e-waste. They contain a substantial number of metals, including precious metals. The increase in consumption of these types of electronics in recent years also indicates an increase in the volume of waste from the use of these types of electronics. Recycling of these types of waste has become a necessary and mandatory field.

The safe and sustainable disposal of e-waste at the end of its use cycle has been seen as a matter of concern for both governments and the public due to its impact on the quality of life of the population and the safety of the environment. The elimination of a heterogeneous mixture of organic materials, metals, etc., involves a special approach and treatment to prevent the inhabitant's exposure to the consequences of the damage resulting from their leakage and dissipation for an effective attenuation of emerging risk phenomena.

In 2019, a record 53.6 million tons of electronic waste such as mobile phones, computers were reported, increasing by 21% in the last 5 years. The Global Waste Monitoring Report predicts that by 2030 the amount of e-waste could reach 74 million tons, double that of 2014 (Forti *et al.*, 2020; e-waste in the EU): data and figures (infographic), 2020).

Printed circuit boards (PCBs) are a major component of electronic remains. Typical PCB composition: non-metals (plastics, epoxy resins, glass) > 70%, copper ~ 16%, solder alloys ~ 4%, iron, ferrite ~ 3%, nickel ~ 2%, silver 0.05%, gold 0.03%, palladium 0.01%, others (bismuth, antimony, tantalum, etc.) < 0.01%. With an estimated over 4 billion phones worldwide and the advancement of technology, a rapid increase in the amount of waste produced by used mobile phones is expected (Zhiqiang *et al.*, 2019).

Recovering precious metals by recycling e-waste is a process 13 times cheaper than extracting metals from mines. Other metals such as copper, nickel and aluminum have a high value and are as recyclable as any other metal.

According to Eurostat, Romania was in last place in the European Union in 2016 for the collection of electrical waste. First place is occupied by Sweden, with 16.5 kg per inhabitant, followed by Great Britain (14.8 kg) and Denmark

(12.4 kg) In terms of recycling this type of waste, in first place is Croatia (81.3%), our country occupies the penultimate place, with a recycling rate of only 25% (European Parliament, Electronic waste in the EU: data and figures, 2020).

Radiographic materials

Another source of precious metals, in fact of silver, is represented by photographic films and materials used in their processing. One of the most useful techniques for diagnosing patient problems, widely used so far is X-ray radiography. In fact, we are only talking about radiological films, because they represent the majority. The corresponding radiographic films and effluents contain significant amounts of silver, for example one kilogram of radiological film may contain between 5 and 15g Ag, representing a valuable source for obtaining this precious metal. Radiological films contain about 7.5 grams of silver / kg, the films used in conventional cameras have around 2 grams Ag / kg and the solutions used for processing have on average about 3 grams of silver / L (depending on the type of solution and processing) (Satyanarayana *et al.*, 2020; Syed, 2016). World-level research to recover high-purity silver from waste using this technique has led to the discovery of simple, fast, inexpensive, and low-pollution methods.

The photosensitive films for radiographs consist of a cellulose nitrate support on which a thin layer of silver halide was deposited on both sides. The approximate composition of X-ray films is as follows: plastic 60%, adhesive layer 3%, emulsion consisting of gelatin and silver halide 25%, supercoat (toxic metal) 10% (Satyanarayana *et al.*, 2020).

About 18-20% of the world's necessary silver is obtained from recycling of photographic waste. Global demand for silver is steadily rising, from 25,700 tons in 2016 to 27,000 tons in 2019. More than half of the world's produced silver is used in industry, minimizing the costs and environmental risks of silver mining ores.

Significant amounts of gold and silver are also recovered from waste processing derived from the jewelry industry (Burat *et al.*, 2019).

Determination of waste composition

Before starting a recycling process, the chemical composition of the waste that is presumed to contain precious metals must be determined. Also, waste must be evaluated in terms of risks to humans or the environment. Information on the composition of electronic waste can be obtained from both the manufacturer's safety data sheet and by taking samples and analyzing them. If these data are not known, further additional steps are needed to continue recycling the waste.

Once the (approximate) chemical composition has been established, it must also be checked whether hazardous substances or organic pollutants are present in this waste. Harmful organic pollutants that may be present in e-waste

are: chloro-phenyl ethane, polychlorinated biphenyls, hexabromobiphenyl, pentachlorobenzene, hexachlorobenzene, etc. (Burat *et al.*, 2019; Fritz *et al.*, 2020; Purchase *et al.*, 2020).

However, both the hazardous substances and the organic pollutants may appear in e-waste in limit concentrations that are set out in a hazardous substances file.

There are many methods to determine the presence of precious metals in waste, such as analytical techniques involving UV-Vis absorption spectrometry, atomic absorption spectroscopy, electrochemical or combined methods (Chen *et al.*, 2022; Hajinia and Heidari, 2021; Hussein *et al.*, 2020; Pourreza and Rastegarzadeh, 2001; Tavakoli *et al.*, 2008).

Regarding the composition of the waste, the materials contained in the waste electronic equipment are:

- Precious metals such as gold, silver and palladium and to a lesser extent platinum and ruthenium;
- Ferrous materials;
- Non-ferrous metals: Cu, Al, Ni, Zn, Sn, Co, In and Ga;
- Non-metals and metalloids such as Se and Te;
- Toxic elements: Hg, Be, Cd, As and Sb;
- Halogens: Br, F and Cl;
- Other substances such as: organic compounds, plastics, glass materials and ceramics.

5. Gold

The most precious, well-known and valued metal, since antiquity is gold. It has sentimental, cultural and financial value, and most people buy gold for various reasons, which are generated by traditional socio-cultural factors, local market conditions and general economic factors.

The demand for gold continues to grow year by year (Fig. 1). The large amount of recycled gold is due to the high number of applications it has, the high market price, but also to the value attributed to it during the international political and economic crises.

Gold for the jewelry industry

The highest annual demand for gold is for jewelry. It has fallen in recent decades, but still accounts for about 50% of total gold demand. India and China are the largest markets, in terms of volume, together accounting for over 50% of current global gold demand. Markets in Asia and the Middle East are dominated by the demand for gold with a high level of purity.

Gold for investment

Gold has unique properties in the investment market, representing a valuable currency. The annual amount of gold traded by investors has increased rapidly (over 200%) in the last 30 years.

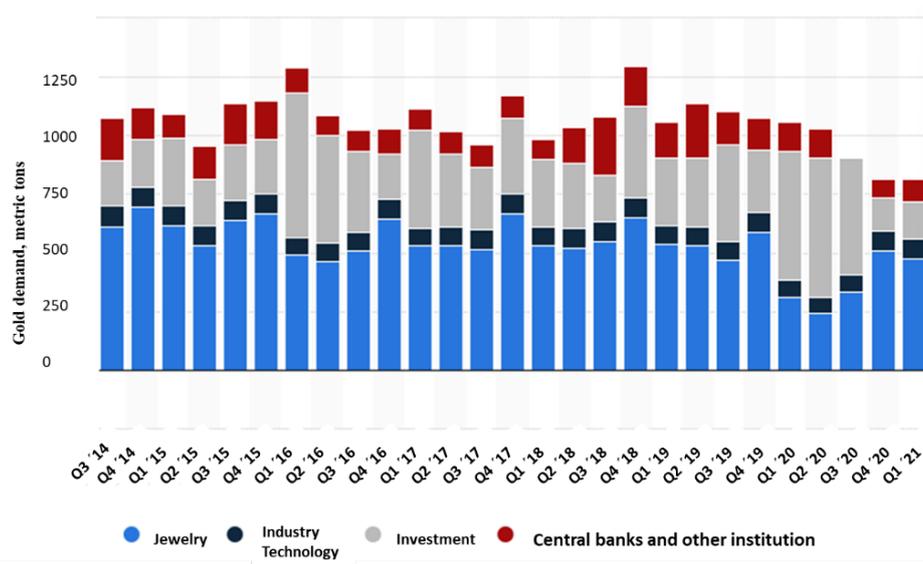


Fig. 1 – Global gold demand from the first quarter of 2016 to the first quarter of 2021, represented by purpose (adjusted by <https://www.statista.com/statistics/274684/global-demand-for-gold-by-purpose-quarterly-figures/>).

Gold demand for banks and financial instruments

In the last 10 years, there has been a major change in the behavior of banks towards gold, a change generated by the relevance of this precious metal in the context of the financial crisis of 2008. Central banks in the domestic markets have increased their official gold purchases, and European banks have reduced or stopped selling, and there is now a high demand for gold.

Gold in industry and technology

An essential factor in the development of the electronics industry was gold, due to its excellent physical properties in this industry. Today, the unique properties of gold (physical and chemical stability, electrical conductivity, optical properties) as well as the development of nanotechnology have led to the discovery of new uses in medicine, engineering and even environmental management. Gold can be used to develop highly accurate methods of administering drugs to the human body, to create conductive compounds and specialized pigments or advanced catalysts that can purify water or air. Since ancient times, gold has been used as a dental material. Although almost all technology applications use small amounts of gold, their impact is very diverse and far-reaching (<https://www.gold.org/about-gold/gold-demand/sectors-of-demand>).

Gold production

Gold production through mining processes accounts for about 75% of annual demand. Mining gold production is lower than demand, so gold shortages need to be met through recycling processes. But gold mining and related activities are relatively slow and do not respond to rapidly changing price changes in the market. Under normal circumstances, there is a long time between exploration and finding new gold deposits and mines that go into production. Because it is practically recyclable indefinitely, almost all extracted gold is, in theory, still accessible in one form or another and may be available for reuse. Recycling is the source of gold supply that responds quickly to the price of gold and economic shocks. About 90% of the recycled gold comes from jewelry, and the gold extracted by other technologies provides the remaining 10%. Gold with guaranteed quality is obtained through purification and refining processes.

6. Recovering / recycling gold

Gold recycling takes place in two stages, the first stage being the generation of a metal concentrate by mechanical enrichment, the second stage being the pyrometallurgical or hydrometallurgical processing of the gold concentrate.

Pyrometallurgical processes have been used for over 30 years and involve incineration, melting of gold-containing materials in a high-temperature furnace. Impurities are removed by various methods (volatilized by a chemical reaction or transformed into slag that rises to the surface of the molten metal or chemically transformed into sludge that is sinking to the bottom). Pyrometallurgy involves a number of processes such as incineration, plasma arc melting, synthesis, melting and gas reactions at high temperatures. More than 70% of electrical and electronic waste is recycled by pyrometallurgy, instead of mechanical processing. The disadvantages of pyrometallurgical processes are obviously represented by the very high energy consumption.

Hydrometallurgical processes

The most important steps in hydrometallurgical processing are represented by a series of acidic or basic leaching of materials containing gold concentrate. Separation and purification procedures are applied to the solutions, for example impurities are precipitated with different reagents (optimally chosen), solvent extraction, adsorption and ion exchange to separate and concentrate the target metals. The solutions are then submitted to electrochemical processes, cementation, chemical reduction or crystallization to recover gold.

Bio-hydro-metallurgical processes

Bio-hydro-metallurgy is a major technology in the materials and minerals industry. The biochemical processes involved in the treatment of metals have

been intensively studied for the last 20 years. Two main classes of biohydrometallurgy process for gold recovery are known: bio-oxidation and bio-sorption. Bio-oxidation is effective for the recovery of gold from metal sulfides, which are the main minerals that have gold, and gold from electronic materials, used bacteria-assisted reactions. Gold recovery is achieved in significant proportions in this way (Achinas *et al.*, 2017; Morin *et al.*, 2006). The bio-absorption process consists in the passive physico-chemical interaction between the surface groups charged with microorganisms and ions in solution. any microorganisms such as algae, bacteria, yeasts and fungi can accumulate gold through various processes. Biosorption processes also have a high efficiency in detoxifying effluents, ensuring multiple advantages.

Fig. 2 shows a general scheme for the processing of electronic waste in order to recover a large percentage of materials.

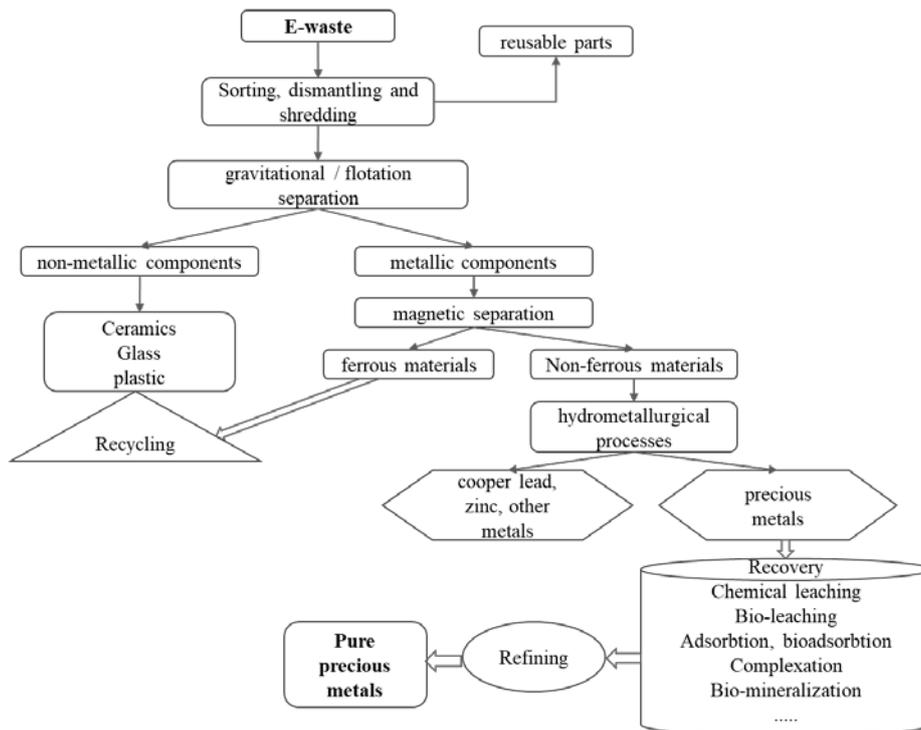


Fig. 2 – Block diagram for the recovery of precious metals from electronic waste.

The precious metal concentrates obtained by various procedures for the processing of electrical and electronic waste, gold and other precious metals can be separated by several physical, chemical, bio-chemical or combined methods, some of them are detailed below, the newest of them being presented in Table 1:

Table 1
*List of current technologies proposed for the recovery of gold
and silver from used materials*

Techniques	Recovery efficiency	Reference
“Selective acid leaching of connector pins” (in H ₂ SO ₄ -H ₂ O ₂ , HCl-H ₂ O ₂ , HCl);	99% for gold	Gontijo and Majuste, 2020
“Hydrometallurgical treatment for activated carbon ash (ACA) via synergistic dual-lixiviant treatment” (thiourea and thiocyanate)	89% Au in 100 min	Adams <i>et al.</i> , 2020
“A cross-linked polymer inclusion membrane (CL-PIM) incorporating the extractant trihexyltetradecylphosphonium bis(2,4,4-trimethylpentyl) phosphinate (Cyphos_ IL 104)”	72-94%	Hoque <i>et al.</i> , 2021
“Chlorate/HCl/CS leaching and bioadsorption of gold ions using GCC beads (0.2 M chlorate, 2 M HCl, 1% chitosan (v/w), a reaction time of 1 h, and a leaching temperature of 55 C)”	≈ 100%	Bui <i>et al.</i> , 2021
“MMT (1-methyl-5-mercapto-1,2,3,4-tetrazole) impregnated activated carbon (MMT-AC)”, reagents for desorption: thiocyanate, thiourea, CN ⁻ , HCl, HBr, thiosulfate	≈ 100% (for thiosulfate desorption)	Chen <i>et al.</i> , 2020
“Emulsion liquid membrane method (organic phase (kerosene, Span80 and MIBK) and internal phase (5% ascorbic acid solution) 1:1)”	high	Zhou <i>et al.</i> , 2021
“Eutectic capture process”	High (Ag, Pd)	Liu <i>et al.</i> , 2020
cellulose-based bio-adsorbent (dithiocarbamate-modified cellulose)	Au ^{III} or Pt ^{IV} , ≈ 99%	Biswas <i>et al.</i> , 2021
“Desorption efficiencies of gold bound COP-180 (A porous porphyrin polymer) treated in HCl-HNO ₃ , thiourea and thiosulfate-sulfite solution”	up to 97%	Son <i>et al.</i> , 2020
Gold adsorption on resorcinol-formaldehyde-biuret (RFB)	High	Xiang <i>et al.</i> , 2020

Hydrometallurgical processes for obtaining precious metals can be accomplished in several variants: leaching of gold with cyanides, halides, thiourea, thiocyanate, thiosulfate, ozone in dilute chloride media, in the chloride-hypochlorite system, with N, N'-disubstituted thiourea, or by bio-oxidation of gold. Gold can be recovered from various leaching mixtures by cementation, adsorption, solvent extraction, ion exchange extraction, coal-oil agglomeration, biosorption - desorption and biomass recovery.

Gold recovery by leaching with cyanide solutions

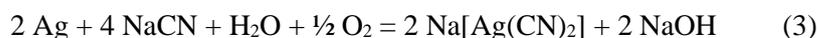
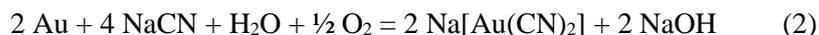
The use of cyanide in mining is old, with sodium cyanide having the property of separating the gold and silver atoms from the rest of the ore. At

present, the methods of using and neutralizing this reagent are very well known, so that there are no risks for humans and the environment.

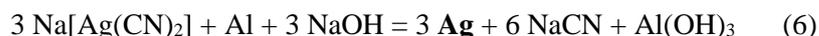
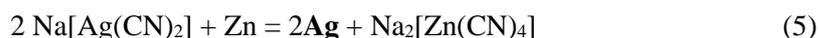
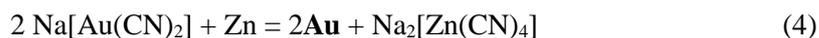
Sodium cyanide has been noted as an active reagent in leaching processes for obtaining gold from primary sources (ores) and from secondary sources (waste). Due to its high efficiency and relatively low cost, more than 18% of total cyanide production is used for gold recovery. In the leaching process, dilute solutions of sodium cyanide (100-500 ppm) are used, the active component that solubilizes gold (through complexing reactions) being cyanide ions (CN^-). The electrochemical process of dissolving gold in cyanide solution is shown in equation (1):



The finely ground waste is mixed at room temperature with an aqueous solution of sodium cyanide alkalized with lime - cyanide attack leaching solution - containing 0.03 - 0.15 g NaCN / 100g and lime 0.05 - 0.5 g CaO / 100g. In the resulting turbidity solution, air is bubbled for 24 hours, when the dissolution of Au and Ag takes place, with the formation of hydrocyanic complexes, soluble in aqueous medium, described in equations (2)-(3):



The liquid phase is separated from the solid phase by decantation and then the recovery and extraction stage of gold and silver is started. Cyanide lye is passed through filters containing zinc or aluminum powder when the reduction process takes place (equations (4) - (6)):



However, a number of environmental accidents around the world have caused widespread concern about the use of cyanide as a leaching reagent. In most of these cases, the cyanide from the processing operations penetrated the environment either through leaks determined by cracks of basins or from storage areas and caused severe contamination of rivers and groundwater. These facts caused widespread concern on the use of cyanide leaching (Gautam *et al.*, 2021).

Gold recovery by leaching with royal water (Aqua regia) and halide solutions

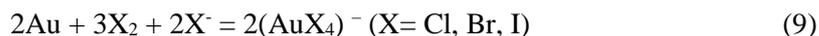
In most hydrometallurgical gold recovery processes, elemental gold is solubilized by oxidation gold ions and then by cyanide complexation.

Royal water (Aqua regia) has a mixture of concentrated nitric acid and hydrochloric acid (1 HNO₃: 3 HCl). The dissolution of gold into royal water is quick, simple and straightforward, but some reaction products (NO_x) are effectively a possibility of air pollution. Equations (7) and (8) describe the formation of trivalent gold ions (Sheng and Etsell, 2007):



Generally, royal water dissolves most metals. Silver has a chemical resistance to royal water. The stability of silver in this mixture is based on passivation. This behavior is due to the spontaneous formation of a thin layer of AgCl and the inner layer can be refined by conventional purification.

Equation (9) describes the dissolution of gold by leaching with halides (fluorine, chlorine, bromine, iodine) processes applied before cyanide:



With the exception of fluorine and astatine, all halogens were tested and used for gold extraction (Syed, 2012). Chlorine, the first halogen used industrially in leaching gold from ores and waste forms with gold forms both complex combinations Au (I) and Au (III), depending on the reaction conditions. Bromine and iodine are chemically similar. Chlorination rates are higher at low pH, high levels of chlorine and chlorides, high temperatures and large specific surfaces. Although the complexation of gold with the chloride anion is much faster than with cyanide, low concentrations of sulfides or other reactive components in the ore can lead to high reagent consumption and reduce the [AuCl₄]⁻ ion - back to metallic gold.

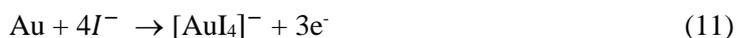
The bromine-bromide system was first recognized as a potential solvent for gold in 1846; however, despite the fact that it has been recognized for a long time, as a powerful gold mining reagent, it has only recently been considered and studied. The advantages of using bromine are short extraction time, lower toxicity, flexibility of pH values. But although the bromine-bromide system is more convenient in many respects compared to cyanide, high reactivity consumption is a major disadvantage. Also, because bromine can react with other elemental species to form toxic compounds, the prices for materials resistant to the severe reaction conditions and the impact on human health can be exorbitant. However, the bromide-bromine system has developed and is widely used, especially since the process of leaching gold based on bromine has been patented. The process is based on a dibromo-dimethyl hydantoin compound (Syed *et al.*, 2012). High extraction yields are obtained using combined processes. Byoung *et al.* (2009) developed an efficient process for recovering gold from waste

materials (electronic waste) by dissolving waste containing gold in royal water at temperature, using a special reactor and dibutyl carbitol (DBC) as extraction reagent. Gold extraction in DBC has been optimized, leading to an efficiency of about 99%. Very small amounts of other elements (Pt, Pd, Ag, Cu and Zn) were extracted in the DBC phase by this process (Byoung *et al.*, 2009).

Gold recovery by leaching in iodine-iodide solutions

The rate of gold leaching in iodine-iodide solutions is much higher than in cyanide processes. Gold leaching in iodine-iodide solution was intensively researched in the 1980s; however, due to high costs, these techniques have not been applied in industry. Nowadays, as the disadvantages of other hydrometallurgies, such as royal water or cyanide, appear, mining and exploration companies are turning their attention to iodide leaching because it has a fast kinetics, easy reaction conditions and the liquid is easy to recover (Liang, 2019).

The anodic reaction of the formation of the gold complex is determined by the iodide ions in the aqueous solution. Because elemental iodine is soluble in water, the oxidation process is limited by I_2 activity. In the presence of free iodide ions, water-soluble complexes are formed – $[AuI_2]^-$ and $[AuI_4]^-$ (equations (10)-(12)).



Gold recovery by leaching with thiourea solution

Thiourea ($(NH_2)_2CS$) is used as a gold extractor for primary and secondary sources. Under acidic conditions, thiourea reacts with gold, forming a cationic complex; the reaction is fast and up to 99% gold can be extracted. The anodic reaction follows equation (13):



However, thiourea must be used under relatively special conditions, as it is quite thermodynamically stable in acidic and neutral media, but decomposes rapidly in basic solutions. Thiourea leaching is effective at $pH = 1-2$ and dependent on a careful control of pH , thiourea concentration and leaching time. Compared to cyanide, thiourea has some advantages, such as low sensitivity to some metals (Pb, Cu, Zn); lower toxicity, low sensitivity to residual sulfur; leading to high recovery of gold from concentrates. Also, the ecological risks are much lower. Industrial use has been reduced in fact because thiourea is more expensive than cyanide; the consumption of thiourea in gold is high for the processing of thiourea is slightly oxidized in solution; and the recovery time is longer.

Gold recovery by leaching with thiocyanate solution

KSCN, potassium thiocyanate is a pseudohalogen that, under certain conditions, forms insoluble salts or complexes with silver, mercury, lead and copper ions and soluble gold-stable soluble complexes. Ion $(SCN)^-$ is an ambidentate ligand (ie it can bind through different atoms to form different coordination compounds) and has an electron donor function either through the sulfur atom or through the nitrogen atom. The SCN^- species can also link two ($M-SCN-M$) or even three metals. Leaching of gold concentrates with acidic potassium thiocyanate solutions produces two complex combinations, $[Au^I(SCN)_2]^-$ and $[Au^{III}(SCN)_4]^-$.

The thiocyanate ion is less toxic, has high stability, but also a low leaching rate. To increase the leaching rate, it is used together with the Fe^{3+} ion, according to equations (14)-(15):

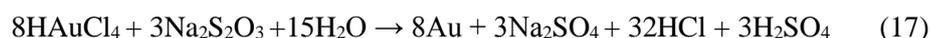


The addition of iron(III) ions optimizes the yield of gold, but the addition of oxygen is not a good option, because the dissolution of gold will be very slow.

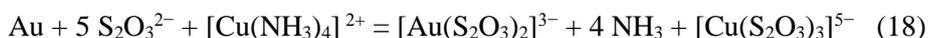
It has been found that the reaction mechanism of gold with potassium thiocyanate in ferric sulphate solutions is directly related to the oxidation of thiocyanate by reducing Fe^{3+} to Fe^{2+} . The oxidation of SCN^- occurs through the formation of several intermediate species, especially trithiocyanate $(SCN)_3^-$ which reacts both as an oxidant and as a complexant for gold. Leaching of gold with thiocyanate, is performed at pH 1-2. More than 95% of the gold is recovered from weak acid solutions for the KSCN concentration of 0.4 mol / L (Azizitorghabeh *et al.*, 2021).

Recovery of gold by leaching with thiosulphate solution

An environmentally friendly alternative to cyanide leaching is thiosulphate solutions. This is done by solubilizing Au and Ag, usually in alkaline conditions, to avoid the breakdown of thiosulphate in the presence of oxygen as an oxidizing agent (equations (16)-(17)).



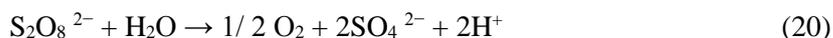
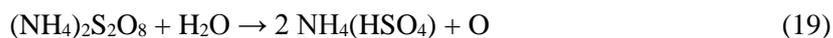
In general, gold reacts more slowly with thiosulfate (in an alkaline medium), but this can be increased by the presence of ammonia and Cu^{2+} ions. Thiosulfate and copper complexes are sensitive to the pH values of the solution. The ammoniacal copper thiosulphate complex dissolves gold according to equation (18):



Thiosulfate leaching can be considered a non-toxic process. The gold extraction yield is close to that obtained by cyanide leaching process but thiosulfate is much cheaper than cyanide. Compared to other methods, the important advantages of leaching with $(\text{S}_2\text{O}_3)^{2-}$ are: low toxicity, possibility to reuse / recirculate leaching reagents, good selectivity, lower reagent price, possibility to recover dissolved gold by adsorption processes or electrodeposition. However, the major disadvantages of thiosulphate leaching in the basic environment are the high volumes of reagents involved and the low extraction yields.

Recovery of gold by leaching with persulphate solution

Syed (2006) reported the process of recovering gold from waste materials by leaching with potassium/ammonium persulphate, according to the following chemical reactions as it is shown in equations (19)-(20):



The process involves the formation of oxidizing ions (SO_4^{2-}) and of active oxygen from $(\text{NH}_4)_2\text{S}_2\text{O}_8$. Then, (O_2) formation by thermal decomposition and the its rapid transfer and dissolution in the aqueous system by the partial increase of pressure follows.

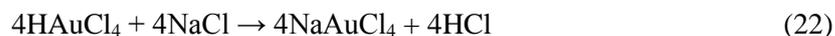
Recovery of gold by leaching with ozone in an acid medium

Aqueous ozone may be a reasonable alternative to leaching concentrates and wastes containing precious metals, mainly due to the formation of oxygen as a reaction by-product. Another advantage is that ozone can be used at a very low aqueous concentration ($\sim 10^{-4}\text{M}$) by injecting O_2 / O_3 mixtures at a low partial pressure of ozone ($P_{\text{O}_3} < 10 \text{ kPa}$). The major disadvantage is electricity consumption, 12–18 kWh / kg O_3 (Viñals *et al.*, 2006).

Ozone bubbling in dilute HCl solutions was used to damage the gold from metal scrap at ambient temperature, the gold dissolves in the $\text{O}_3 / \text{Cl}^- / \text{H}^+$ system with the formation of $[\text{AuCl}_4]^-$, the procedure can be applied effectively for recovery palladium and rhodium in waste (Viñals *et al.*, 2006).

Recovery of gold by leaching in the chloride-hypochlorite system

Leaching in the chloride-hypochlorite system is an alternative way to obtain gold. The reactions between gold and chloride-hypochlorite leaching solutions with $[\text{AuCl}_4]^-$ complex ion formation are presented below (equations (21)-(23)):



The general equation can be written as (equation (24)):

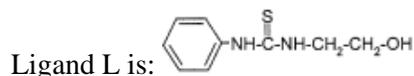
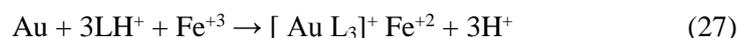
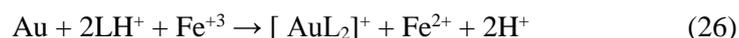


With a yield of about 90% (for a reaction time of 30 minutes) pure gold was obtained by leaching with acidic NaClO solutions, the addition of HClO being able to reduce the amount of sodium hypochlorite used. Better extraction yields were obtained by increasing the temperature or replacing NaClO with Ca(ClO)₂. Under the same conditions, silver was extracted, with a yield higher than 95%, a yield that for silver cannot be obtained in the classic oxidation-cyanide processes (Hasab *et al.*, 2013).

Gold recovery by leaching with N, N'-disubstituted thiourea

1-Phenyl-2-thio-3- (2-hydroxyethyl) urea (thiourea N, N'-disubstituted), in the presence of an oxidant (such as Fe³⁺ / H₂SO₄) dissolves the gold at room temperature, with a yield above 95%. The experimental results obtained in this (Aydin *et al.*, 2001) showed that open chain substituted thiourea derivatives can function as very good leaching agents.

The general equations for gold reaction with an oxidant in the presence of N, N'-disubstituted thiourea acting as a complexing reagent, are given below as equations (25)-(27):



According to the above equations, metallic gold is oxidized by Fe(III) ions in the presence of N, N'-disubstituted thiourea reagent, and the cationic complex combinations [AuL₂]⁺ and [AuL₃]⁺ are transferred into solution (Aydin *et al.*, 2001).

Gold recovery through bio-leaching and bio-oxidation processes

The recovery of valuable metals from various wastes using microbial species has attracted much interest in recent years due to the long-term decline in ore reserves and concentrates. Microorganisms play an important role especially in the gold recovery process. Some specialized bacteria, fungi, yeasts, algae or

actinomycetes are being used more and more to facilitate the extraction of gold from low-grade gold concentrates. These microorganisms can increase the oxidation of metallic minerals and can be used as flotation agents or as biosorbents in the gold recovery process.

In recent years, two different types of bio-obtaining processes have attracted the interest of researchers as alternatives to conventional methods: bioleaching and biological oxidation, as they have proven to be cost-effective, sustainable and non-hazardous (Rana *et al.*, 2020).

Bio-leaching is a solubilization process in which bacteria are very helpful in dissolving gold from ores or waste, while in the bio-oxidation process, acidophilic microorganisms release gold from minerals during sulfides oxidation (Johnson, 2014). *Static bio-oxidation techniques are based on the principle of water and air circulation through piles of waste in order to activate the growth of microorganisms that amplify oxidation.* Cyanogenic microorganisms such as *Chromobacterium violaceum*, *Pseudomonas fluorescens* and *Pseudomonas plecoglossicida* have led to the recovery of gold when grown in metal-containing culture media. Compared to chemical oxidation, biological oxidation offers the advantages of low production costs, low temperatures, low pressures, partial sulfides oxidation, low leaching reagent consumption and the absence of air pollution (Kaksonen *et al.*, 2014). Several acidophilic bacteria, iron chemolithotrophs and oxidizing sulfur are present in polymetallic sulfide ores. Mesophilic oxidizing bacteria of iron and sulfur such as *Acidithiobacillus ferroxidans*, *Acidithiobacillus thiooxidans* oxidizing sulfur, *Leptospirillum ferriphilum* and *Leptospirillum ferrooxidans* oxidizing iron, moderately thermophilic bacteria such as *Acidithiobacillus oxidus* were related as helpful to sulphides oxidation. Various microorganisms have been tested for gold bio-leaching (Kaksonen *et al.*, 2014).

Bio-sorption by microbial biomass is another promising, low cost and environmentally friendly method for recovering gold from waste. Bio-sorption is a passive method of absorption and / or complexation of gold in the cell wall of various biomasses of algae, fungi and bacteria (Mata *et al.*, 2009). Stationary or dead microbial biomass binds and concentrates gold ions in the leaching concentrated solutions. In the bio-sorption processes, a series of green and brown algae (*Chlorella vulgaris*, *Fucus vesiculosus*, *Sargassum natans*), fungi (*Aspergillus niger*, *Mucor rouxii*, *Rhizopus arrizus*, *Aspergillus oryzae*, *Chaetomium globosum*, *Gibberella fujikurium*, *Penicium*, *Hyucum*, *Hyucelium*, *Penicium*), yeast (*Candida krusei*, *Candida robusta*, *Candida utilis*, *Cryptococcus albidus*, *Cryptococcus laurentii*, *Debaryomyces hansenii*, *Endomycopsis fibuligera*, *Hansenula anomala*, *Hansenula saturnas*, *Kluyveromyces spp.*, *Saccharomyces cerevisiae*, *Sporobolceses calcoaceticus*, *Erwinia herbicola*, *Pseudomonas aeruginosa*, *Pseudomonas maltophilia*) are used for gold recovery (Syed, 2012, 2016).

Recent studies have shown effective results for gold recovery with other bioleaching methods such as bio-precipitation, bio-mineralization, bio-flotation, bio-flocculation, bio-sorption and bioaccumulation (Achinas *et al.*, 2017). All these methods are based on microorganisms necessary for “harvesting” gold, which are either isolated in gold-enriched areas, or as domesticated strains, with exceptional gold recovery efficiency (Jingying *et al.*, 2020; Ju *et al.*, 2016; Rana *et al.*, 2020).

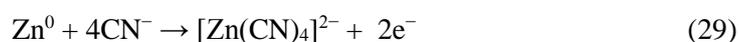
Due to the fact that different microbial communities have different structures, functions and dynamics in gold metabolism, their use in bio-hydrometallurgical processes remains a challenge. Even though many thermophilic or acidophilic bacteria or other types of microorganisms have been isolated, characterized and even used for the precious metals extraction, new strains isolated from various sources of waste are needed. At the same time, it is necessary to find combinations of chemicals compatible with effective microbial reagents to recover high levels of gold (Rana *et al.*, 2020).

In the mining industry, natural microorganisms that can be exploited through various gold extraction and recovery strategies have great potential. *The use of microorganisms for precious metals recovery from waste is economical and avoids environmental pollution.* Moreover, process-based microorganism-based technologies such as biofiltration using specialized biofilters can ensure a specific recovery of gold ions. Future microorganism-based gold recovery processing systems will revolutionize gold production.

For gold recovery from various solutions, the literature offers different methods, such as cementation, chemical extraction of the precipitating solvent, ion exchange, adsorption, bio-sorption, electrolysis, coagulation, etc.

Cementation

The zinc cementation process for recovering gold from cyanide leaching solutions has been known and used since ancient times. The redox reactions that take place are described in equations (28)-(29):



Cementation can also be done with magnesium, aluminum, zinc, iron and copper.

Adsorption

The adsorption technique for recovering gold cyanide complexes from an aqueous solution of activated carbon was first patented by McQuiston and Chapman (1951). Since then, this process has been substantially improved, and now carbon adsorption technologies are being used commercially for gold recovery. The mechanism and kinetics of adsorption of gold from acid solutions

of thiourea on activated carbon have been extensively studied, the adsorption efficiency of gold decreases with increasing thiourea concentration, pH and temperature. The advantage is that the adsorption of gold is not influenced by small amounts of Fe^{2+} or Fe^{3+} , but is strongly reduced by Ag^+ and Cu^{2+} ions, due to competitive adsorption processes (because they compete strongly for surface adsorption positions). The adsorption rate of gold in the initial stage is described by first order kinetics. Activated charcoal used for the adsorption of gold from various mixtures can also be obtained from vegetable calcination residues (rice husk, barley straw, apricot kernels, etc.). Desorption studies were performed with a mixture of aqueous sodium hydroxide solution and organic solvents at ambient temperature (Syed, 2012).

Solvent extraction

Organic solvent extraction is a convenient option for separating and recovering gold from primary and secondary leaching solutions. Various emerging systems have been used around the world to recover gold. The extraction of Au (III) from acidic solutions in the CTAB / n-heptane / iso-amyl alcohol / Na_2SO_3 system was 99% efficient, and was selective for Au (III) ignoring the metal ions of Fe^{3+} , Al^{3+} , Cu^{2+} , Ni^{2+} (Wenjuan *et al.*, 2011).

There are many organic solvent systems (for gold mining such as polyethylene glycol, strongly basic macro-cyclic tri-amines, 1,5,9-triazocyclododecane, mixed primary amine extraction system and tri-butyl phosphate or tri-alkylamine oxide etc.), many of these systems are patented and marketed. There are many organic solvent systems used for gold extraction (such as polyethylene glycol, strongly basic macro-cyclic tri-amines, 1,5,9-Triazocyclododecane, mixed extraction system based on primary amine and tri-butyl phosphate or tri-alkyl amine oxide, etc.). Cyanex 272, many of these systems are patented and marketed (Kubota *et al.*, 2019; Rao *et al.*, 2021; Xing *et al.*, 2018).

3. Conclusions

Electrical and electronic waste is an environmental challenge for which solutions are still being sought. There are many ways in which more or less efficient technologies can be used to process this waste and recover as much materials as possible, with an emphasis on the recovery of precious metals. The costs of processing this waste and recovering the metallic materials are much lower than obtaining the metals by extracting and processing the ores. Regarding the recovery of gold and silver, in recent years the focus has been on adsorption processes using selective adsorbents based on activated carbon or polymer resins, and also on extraction with different solvents or mixed leaching systems, most often combined with bioleaching, bio-sorption, bio-mineralization or bioaccumulation methods.

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- <https://www.statista.com/statistics/274684/global-demand-for-gold-by-purpose-quarterly-figures/>

TENDINȚE ACTUALE PRIVIND GESTIONAREA DEȘEURILOR ELECTRICE ȘI ELECTRONICE CONȚINÂND METALE PREȚIOASE

(Rezumat)

Recuperarea metalelor prețioase se poate face eficient din deșeurile care sunt considerate surse secundare de materii prime, minimizând nevoia de exploatare și prelucrare a materiilor prime, reducând în același timp consumul de energie și apă. Deșeurile electrice și electronice, filmele fotografice uzate sau deșeurile provenite de la fabricarea bijuteriilor se încadrează în această categorie și duc la recuperarea a aproximativ 25% din necesarul anual de aur și 20% din necesarul anual de argint. Lucrarea prezintă avantajele reciclării deșeurilor care conțin metale prețioase, dificultățile procesului de reciclare precum și metodele de recuperare a metalelor nobile prin prelucrarea deșeurilor electrice și electronice, cu accent pe tehnologiile lansate în ultimii ani (procedee bio-hidro-metalurgice, procese de bio-oxidare statică, adsorbție pe cărbune activ sau membrane polimerice, extracție selectivă).