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REVIEW

Gold recovery from primary and secondary raw materials: leaching, matrix effects and selective separation

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ABSTRACT

Gold ore grades are declining, refractory mineralization is increasing, and electronic scrap and copper anode slime have reached industrial volumes. For such feeds, the rate-limiting stage is selective separation from a chemically loaded matrix, whereas dissolution chemistry is established. This review compares cyanide, thiosulfate, thiourea, thiocyanate, halide, glycine, and enhanced-oxidation systems with respect to their applicability to both primary and secondary feeds. Cyanidation remains the benchmark, but its margin narrows once copper consumption, encapsulation, and detoxification costs are taken into account. Each alternative lifts one restriction and imposes others on reagent stability and selectivity. Progress depends on integrating dissolution, separation and effluent management for heterogeneous feeds.

Keywords: gold, hydrometallurgy, lixiviants, selectivity, refractory ores, secondary raw materials

INTRODUCTION

Gold is used in electronics, telecommunications equipment, medical devices, catalysis, aerospace applications and jewelry manufacture. This range of applications is supported by its high electrical conductivity, corrosion resistance, chemical inertness, ductility and biocompatibility [1, 2]. Gold also serves a monetary function. Investment and industrial demand act at the same time and rarely move in phase, which keeps pressure on the raw material base (**Fig. 1**) [3]. Between 2015 and 2025 demand in physical terms remained within a narrow band of about 4,400 to 5,000 t per year, an increase of roughly 13% over the decade, whereas its monetary value rose from approximately US$170 billion to about US$550 billion, with most of that gain concentrated in 2023-2025. Physical volume is therefore close to saturation, while the value of the same volume has more than tripled.

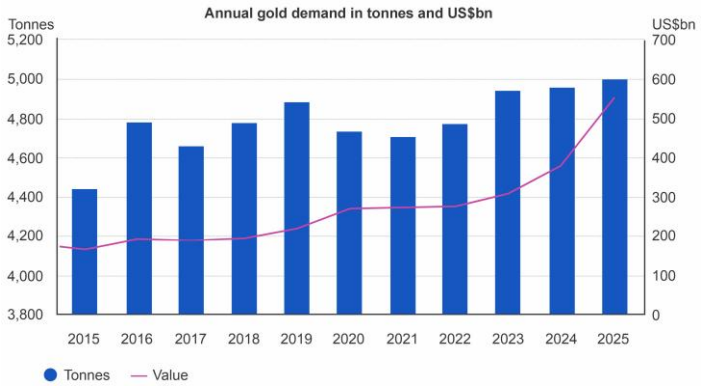

Fig. 1 Annual gold demand in tonnes (bars, left axis) and in US$ billion (line, right axis), 2015-2025 [3].

According to the U.S. Geological Survey, annual primary production remains at the level of several thousand tonnes, whereas economically viable reserves are finite and geographically concentrated [4].The average gold grade of primary ores has been declining for decades, and the share of low-grade, mineralogically complex deposits in the production mix is increasing [5]. A substantial part of the remaining resources is refractory: gold is locked within sulfide minerals, carbonaceous matter or silicate matrices and is not liberated by grinding followed by leaching [6, 7]. Refractory ores require higher energy and reagent input, and the main technological load moves from leaching to pretreatment (**Fig. 2**). The processing routes fall into three groups. Free-milling ore passes from primary crushing, process mineralogy, comminution, classification and physical concentration (gravity separation, flotation) directly to leaching. Refractory and double-refractory ore is first subjected to oxidative pretreatment by roasting, pressure oxidation, biooxidation or ultrafine grinding with atmospheric oxidation, and only then to leaching, which is carried out by cyanidation or with non-cyanide lixiviants (thiosulfate, glycine, thiourea, thiocyanate, halides). Gold from copper and Pb-Zn ores enters as a by-product: the copper route delivers it in anode slime, which after leaching, alkali fusion or smelting joins the main line at the recovery stage, while the Pb-Zn route delivers it in doré bullion, which enters at final refining by the Miller and Wohlwill processes. All routes converge on recovery from pulp or solution (carbon adsorption, resin-in-leach or resin-in-pulp, Merrill-Crowe cementation), electrowinning and refining to 99.99% gold, with leach residues passing to cyanide destruction and tailings storage.

Secondary anthropogenic materials have reached a scale at which they now constitute an independent source of feed. Printed circuit boards and other electronic components contain gold at concentrations appreciably higher than those of many primary ores, while copper anode slime, the residue of copper electrorefining, concentrates precious metals as a by-product of base metal production [2, 8, 9, 10, 11]. Primary ores are geologically constrained, so their mineral associations and impurity distributions are comparatively stable and can be characterized in advance. The composition of secondary materials is not subject to such a constraint: it follows from the design decisions of manufacturers and from market dynamics, with changes from one production year to the next, and is heterogeneous even within a single batch [12, 13].

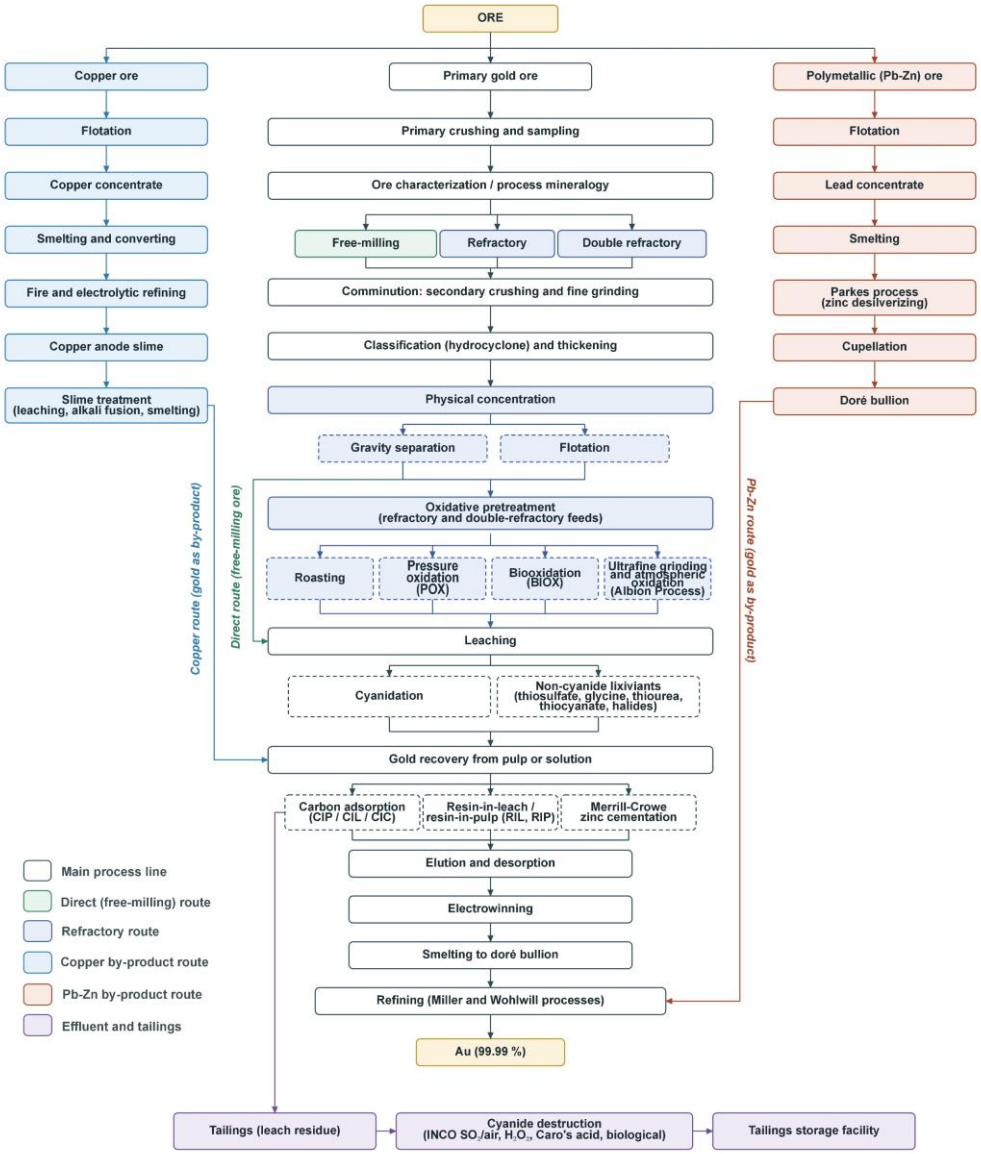

Fig. 2 Processing routes for gold-bearing raw materials: free-milling, refractory and double-refractory primary ores pass through comminution, preconcentration and, where required, oxidative pretreatment (roasting, pressure oxidation, biooxidation) before leaching by cyanidation or by alternative lixiviants**.** Copper and Pb-Zn ores are shown as by-product sources of gold.

Gold dissolution proceeds by the same reaction in an ore and in a milled printed circuit board alike. The difficulty lies in the selective and reproducible separation of gold from a matrix carrying copper, tin, lead, iron, silver, platinum group metals and chalcogens and, for electronic scrap, an organic and halogenated fraction [12].

MATERIAL AND METHODS

Cyanidation as the benchmark and its matrix-dependent limits

Cyanide leaching remains the benchmark process against which alternatives are assessed. In a dilute alkaline solution, typically at pH 10 to 11, gold is oxidized in the presence of dissolved oxygen and complexed into the stable dicyanoaurate ion according to Elsner's equation:

$$4Au + 8CN^- + O_2 + 2H_2O \rightarrow 4[Au(CN)_2]^- + 4OH^- \quad (1)$$

For free-milling ores the process is efficient, robust and scalable, in carbon-in-pulp and carbon-in-leach circuits it consistently provides high recovery [14, 15]. Cyanidation predominates industrially for a chemical reason: the cyanide ligand forms a strong and selective complex with gold under mild and inexpensive conditions.

The efficiency of cyanidation depends on the matrix. It decreases when the feed contains copper minerals that consume cyanide and form competing copper-cyanide complexes, when carbonaceous matter adsorbs the already dissolved gold complex (preg-robbing), and when gold is encapsulated in sulfides and does not contact the solution [16, 17]. All three failure mechanisms originate in the matrix, and all three become more pronounced as feeds shift toward refractory ores and toward secondary materials rich in base metals.

The environmental and regulatory status of cyanide compounds complicates their application to secondary feeds. The toxicity of cyanide and the risk associated with tailings management have led to restrictions or bans in several jurisdictions and to strict regulation elsewhere [18, 19, 20, 21]. In electronic scrap processing the pregnant solution also carries dissolved base metals and organic degradation products, so the effluent load is higher than in ore processing and subsequent detoxification becomes a significant cost item. Oxidative destruction, complexation or biological treatment of cyanide in gold plant residues forms part of any cyanide flowsheet applied to complex feeds, and its cost should be taken into account when weighing the dissolution advantage [22].

Refractoriness and the pretreatment burden

Where gold is enclosed in a sulfide matrix, no lixiviant gains access to it without prior liberation, so the oxidative pretreatment selected governs the economics of the operation as a whole. Three routes are applied industrially.

Roasting oxidizes sulfides at 500 to 700 °C, producing a porous calcine that exposes gold to subsequent leaching [23, 24]. The route is effective but generates sulfur dioxide and, where arsenic is present, volatile arsenic compounds, and therefore requires gas capture and stabilization systems that increase capital and operating costs.

Pressure oxidation decomposes sulfides in an autoclave, usually at 180-230 °C and elevated oxygen partial pressure, retaining sulfur and arsenic predominantly in the aqueous phase rather than releasing them into the off-gas [25, 26]. The route provides deep sulfide oxidation and high subsequent recovery but requires substantial capital investment and the use of corrosion-resistant materials of construction. Gold recovery after cyanidation of pressure-oxidized refractory feeds is reported to exceed 95% [27, 28]**.** The value depends on mineralogy and process conditions, so data from individual studies are indicative only.

Biooxidation employs iron- and sulfur-oxidizing microorganisms to decompose the sulfide matrix under mild conditions:

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O \quad (2)$$

$$FeS_2 + 6Fe^{3+} + 3H_2O \rightarrow S_2O_3^{2-} + 7Fe^{2+} + 6H^+ \quad (3)$$

$$FeAsS + 11Fe^{3+} + 7H_2O \rightarrow 12Fe^{2+} + H_3AsO_3 + SO_4^{2-} + 11H^+ \quad (4)$$

The route is energy-efficient and free of the gas emissions associated with roasting**.** ***It*** has been applied to low-grade and arsenic-bearing refractory ores [29, 30, 31]. Its limitations are kinetic: residence times are long and microbial activity is sensitive to feed chemistry, which reduces throughput and complicates the treatment of variable material [32, 33].

All three routes add a separate, cost-bearing stage prior to gold dissolution. For refractory primary ores this stage dominates the cost structure. For secondary materials the pretreatment takes a different form, and its share of the flowsheet increases as the matrix becomes more complex.

Non-cyanide lixiviants and the limits of transferability

The search for alternative lixiviants has been driven by environmental and regulatory pressure associated with the use of cyanide. Each system lifts a specific matrix restriction and imposes new ones [34].

Thiosulfate. Ammoniacal thiosulfate leaching is carried out in a mildly alkaline solution (pH 9.0-10.5) with the copper-ammine Cu(II)/Cu(I) couple acting as the electron carrier. Gold is oxidized not directly by oxygen but by the tetraamminecopper(II) ion:

$$Au + 5S_2O_3^{2-} + [Cu(NH_3)_4]^{2+} \rightarrow [Au(S_2O_3)_2]^{3-} + [Cu(S_2O_3)_3]^{5-} + 4NH_3 \quad (5)$$

Copper(I) is regenerated by oxygen in the ammoniacal medium:

$$4[Cu(S_2O_3)_3]^{5-} + 16NH_3 + O_2 + 2H_2O \rightarrow 4[Cu(NH_3)_4]^{2+} + 12S_2O_3^{2-} + 4OH^- \quad (6)$$

The overall reaction is:

$$4Au + 8S_2O_3^{2-} + O_2 + 2H_2O \rightarrow 4[Au(S_2O_3)_2]^{3-} + 4OH^- \quad (7)$$

Thiosulfate leaching tolerates the matrices that disrupt cyanidation, namely preg-robbing carbonaceous ores and copper-bearing feeds [35, 36]. The system is therefore suited to refractory ores as well as to copper-bearing electronic scrap, and it has been demonstrated on an industrial scale [37]. The limitation is reagent stability. The copper(II) that oxidizes gold also oxidizes the ligand to tetrathionate, with subsequent conversion to trithionate and sulfate:

$$2S_2O_3^{2-} + 2[Cu(NH_3)_4]^{2+} \rightarrow S_4O_6^{2-} + 2[Cu(NH_3)_2]^+ + 4NH_3 \quad (8)$$

Reagent consumption is high, and the copper and ammonia concentrations require precise control, which increases cost and complicates process operation [38].

Thiourea. In acidic solution (pH 1-2) with ferric ions as the oxidant, thiourea rapidly dissolves gold as a cationic complex:

$$Au + 2CS(NH_2)_2 + Fe^{3+} \rightarrow [Au(CS(NH_2)_2)_2]^+ + Fe^{2+} \quad (9)$$

The fast kinetics make the system applicable to high-grade concentrates and to readily accessible gold in electronic scrap [39]. The same Fe^{3+} oxidizes the reagent to formamidine disulfide:

$$2CS(NH_2)_2 + 2Fe^{3+} \rightarrow (NH_2)(NH)C\text{-}S\text{-}S\text{-}C(NH)(NH_2) + 2Fe^{2+} + 2H^+ \quad (10)$$

This step is reversible, but above an Eh of approximately 0.45 V (SHE) the disulfide decomposes irreversibly:

$$(NH_2)(NH)C\text{-}S\text{-}S\text{-}C(NH)(NH_2) \rightarrow CS(NH_2)_2 + NH_2CN + S^0 \quad (11)$$

The elemental sulfur released passivates the gold surface, and reagent loss increases operating costs. The acidic effluent requires separate handling. Reported recoveries are high at laboratory scale, whereas industrial scale-up remains limited [40, 41].

Thiocyanate. In an acidic medium (pH 1-3) with Fe^{3+} as the oxidant, gold is dissolved as Au(I) or Au(III) depending on the potential:

$$Au + 2SCN^- + Fe^{3+} \rightarrow [Au(SCN)_2]^- + Fe^{2+} \quad (12)$$

$$Au + 4SCN^- + 3Fe^{3+} \rightarrow [Au(SCN)_4]^- + 3Fe^{2+} \quad (13)$$

Thiocyanate systems provide high recovery from oxidized ores with relatively fast kinetics and appreciably lower acute toxicity than cyanide [42, 43]. Their application is limited by the high consumption of reagent and oxidant and by kinetics slower than those of cyanide**.** Industrial development remains at an early stage.

Halides. Chloride, bromide and iodide systems dissolve gold in strongly acidic oxidizing media through the formation of tetrahaloaurate complexes:

$$2Au + 3Cl_2 + 2Cl^- \rightarrow 2[AuCl_4]^- \quad (14)$$

$$2Au + 3Br_2 + 2Br^- \rightarrow 2[AuBr_4]^- \quad (15)$$

Iodide systems form a stable gold(I) complex and operate under milder conditions:

$$2Au + 4I^- + H_2O_2 + 2H^+ \rightarrow 2[AuI_2]^- + 2H_2O \quad (16)$$

In electrochemical chloride routes the anodic half-reaction is:

$$Au + 4Cl^- \rightarrow [AuCl_4]^- + 3e^-,\ E^\circ = +1.00\ V\ (SHE) \quad (17)$$

These systems perform well on concentrates and high-grade materials, including electronic scrap [44, 45, 46]. They impose material and safety constraints. Strongly acidic media require corrosion-resistant construction materials, chlorine evolution requires containment, and effluent treatment adds cost.

Glycine. Glycine forms a stable bis-glycinate gold(I) complex in an alkaline medium (pH 10-11, 40-60 °C) in the presence of oxygen:

$$4Au + 8NH_2CH_2COO^- + O_2 + 2H_2O \rightarrow 4[Au(NH_2CH_2COO)_2]^- + 4OH^- \quad (18)$$

In molecular form, using glycine and alkali:

$$4Au + 8NH_2CH_2COOH + 4NaOH + O_2 \rightarrow 4Na[Au(NH_2CH_2COO)_2] + 6H_2O \quad (19)$$

Glycine has low toxicity and can be regenerated. Most published work addresses copper-gold systems, where glycine leaches copper selectively [47, 48, 49]. Its kinetics are slower than those of cyanidation, and the technology remains under development.

Enhanced-oxidation systems. Instead of replacing the complexing ligand, these processes generate reactive radicals. The hydroxyl radical is produced by the Fenton reaction and the sulfate radical anion by activation of peroxodisulfate:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {\bullet}OH + OH^- \quad (20)$$

$$S_2O_8^{2-} + Fe^{2+} \rightarrow Fe^{3+} + SO_4^{2-} + SO_4{\bullet}^- \quad (21)$$

The radicals oxidize metallic gold, which is then complexed by the ligand L^- present in solution (halide, thiosulfate, amino acid):

$$Au + 2L^- + {\bullet}OH \rightarrow [AuL_2]^- + OH^- \quad (22)$$

Published applications to electronic scrap are recent and have been carried out predominantly at laboratory scale [50, 51].The approach targets readily accessible

metallic gold, for example in processor components, and requires neither strong acids nor cyanide [52].

Specific matrix problems of secondary raw materials

The accompanying interferences, which have no close analogue in primary ores, govern the difficulty of processing secondary materials.

The heterogeneity of electronic scrap follows from product design. A single batch may contain base metals at the weight percent level, precious metals at trace and low percent levels, and a substantial proportion of plastics, glass fiber and brominated flame retardants, with the ratios varying with device type and production period [12, 53]. Recovery therefore begins with pretreatment: dismantling, comminution and physical separation, which expose metallic surfaces and remove the organic fraction (**Fig. 3**).

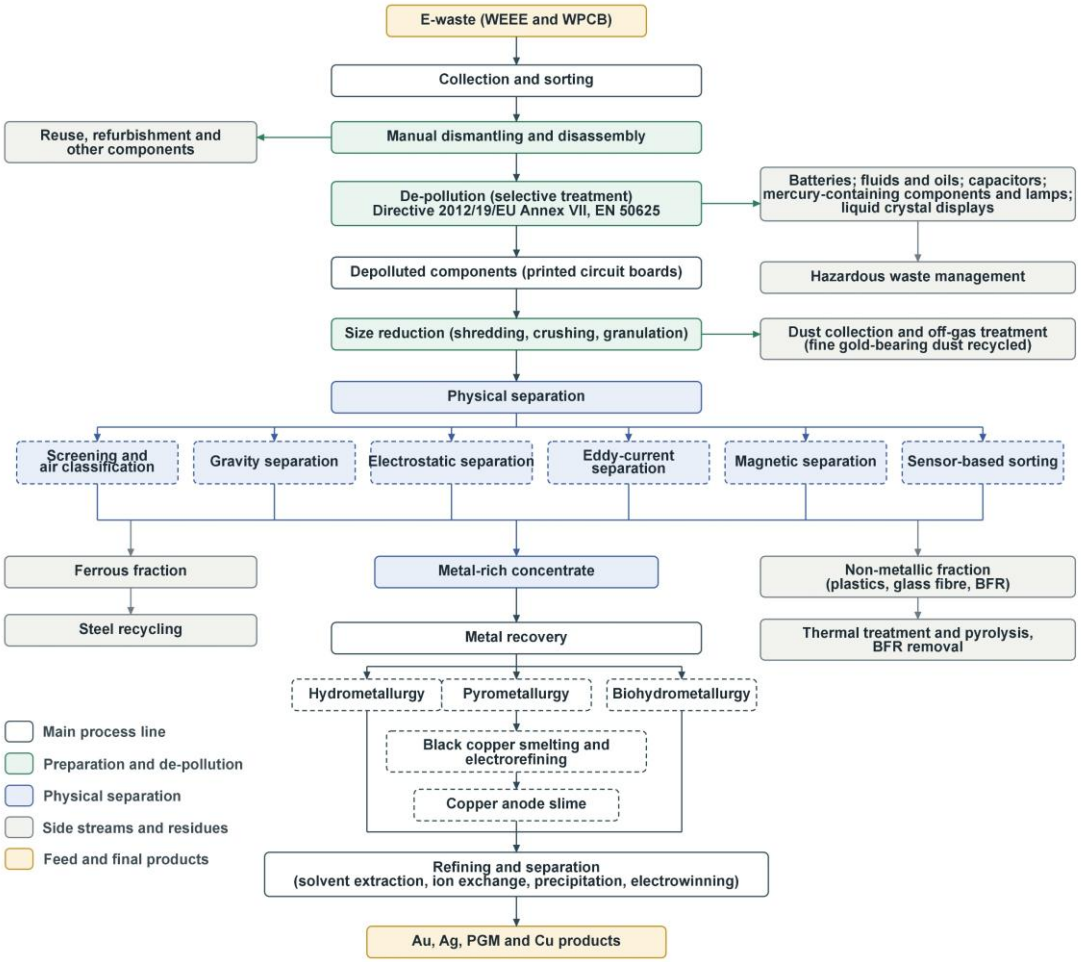

Fig. 3 Preparation and metal valorization sequence for electronic scrap: sorting, manual dismantling with removal of hazardous components, comminution and physical separation (gravity, electrostatic, eddy-current and magnetic) preceding hydrometallurgical, pyrometallurgical or biohydrometallurgical treatment.

The sequence starts with collection and sorting, manual dismantling with the removal of reusable components, and de-pollution in accordance with Directive 2012/19/EU Annex VII and EN 50625, which withdraws batteries, fluids and oils, capacitors, mercury-containing components and lamps and liquid crystal displays into hazardous waste management. The depolluted printed circuit boards are then shredded, crushed and granulated, with fine gold-bearing dust captured in the off-gas system and returned to the circuit, and are passed through screening and air classification, gravity, electrostatic, eddy-current and magnetic separation and sensor-based sorting. Three streams leave this stage: a ferrous fraction to steel recycling, a non-metallic fraction of plastics, glass fibre and brominated flame retardants to thermal treatment and pyrolysis, and a metal-rich concentrate, which is the only stream carrying gold forward to hydrometallurgical, pyrometallurgical or biohydrometallurgical recovery and further refining to Au, Ag, PGM and Cu products. Each of these stages causes irreversible gold losses to the non-metallic, ferrous and dust fractions [54, 55]. In practice, gold is more often recovered from electronic scrap through integrated pyrometallurgical concentration followed by hydrometallurgical separation**.** This route resembles the black copper process in primary production and concentrates gold in the copper-bearing phase before selective dissolution [56, 57, 58].

Copper anode slime is a predominantly inorganic, metal-rich residue in which gold occurs as metallic particles or as a gold-silver alloy, often locked in multiphase aggregates together with selenides, tellurides and base metal phases [59, 60]. Its composition varies widely, and gold contents compiled from plant surveys are reported over a broad range (**Fig. 4**) [9]. Copper dominates the reported compositions, with a median of about 17 wt.% and an interquartile range of roughly 9 to 25 wt.% against a total spread of 0.5 to 41 wt.%. Selenium follows with a median near 8 wt.% and a maximum of about 21 wt.%, silver with a median near 7 wt.% and a spread reaching 33 wt.%, and lead with a median of about 4 wt.% and a spread reaching 32 wt.%. Antimony, arsenic, tellurium and bismuth stay below roughly 5 wt.% in most surveys, while gold itself has a median below 1

wt.% and seldom exceeds 1.5 wt.%, and Pt, Pd, Ni, Sn, Fe and Zn occur only at fractions of a percent. For every major component the spread is comparable to or larger than the median, so a single reported composition cannot be treated as representative and the flowsheet has to be designed for a range rather than for a fixed feed. The increasing share of secondary feed in copper smelting has given rise to so-called non-standard anode slime, in which poorly soluble phases, primarily metastannic acid, form gelatinous or finely dispersed matrices that hinder lixiviant access to gold [13, 61]. The interference here consists of refractory secondary phases which have to be converted before gold can be liberated. Alkali fusion, usually with sodium hydroxide or a hydroxide-nitrate flux, is used to convert these phases into soluble compounds while gold remains in the enriched residue, very high gold retention is reported for optimized fusion-leaching routes [62, 63, 64, 65].

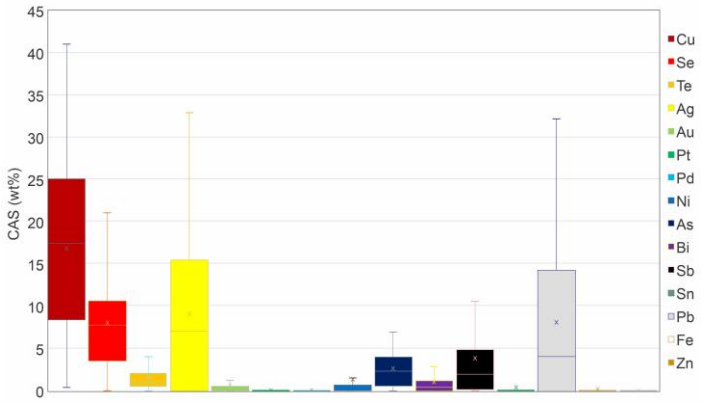

Fig. 4 Elemental composition of copper anode slime from different sources, wt.%: box-and-whisker plot showing the interquartile range, median, mean and total spread of the reported values redrawn from data compiled in [9].

Both secondary materials carry gold at concentrations that would be attractive in an ore, yet their technical obstacles are opposite in nature: dispersion in a heterogeneous, organic-rich matrix in the case of electronic scrap and encapsulation in refractory inorganic phases in the case of anode slime. Improved dissolution chemistry addresses neither obstacle. Both require matrix transformation followed by selective separation.

Downstream selectivity: separation and purification

Once gold has been brought into solution, recovery is completed by separating it from co-dissolved components and reducing it to the metal. For complex feeds, selectivity is most critical at this stage. The principal operations are solvent extraction, adsorption on activated carbon or functionalized sorbents, ion exchange, precipitation and electrowinning, each with its own balance of selectivity, cost and scalability (**Fig. 5**) [8, 66]. The route is selected according to the number of competing metals in the pregnant solution. A binary system, containing gold and a single competitor, passes directly to reduction of gold to the metal by precipitation or electrowinning. A multimetallic solution carrying copper, iron, nickel, silver and palladium requires an intermediate separation stage before reduction, and each option has its own constraint: liquid-liquid extraction imposes organic-phase turnover, ion exchange is sensitive to competing ions, and selective sorbents operate only at low gold concentrations.

For clean solutions the final reduction step is efficient: precipitation or electrowinning under optimized conditions recovers more than 99% of the dissolved gold. Lower values reported for non-optimized systems indicate incomplete reduction [67, 68]. For multi-metal solutions the principal problem is selectivity. Solvent extraction offers high selectivity and scalability but requires management of the organic phase [66].

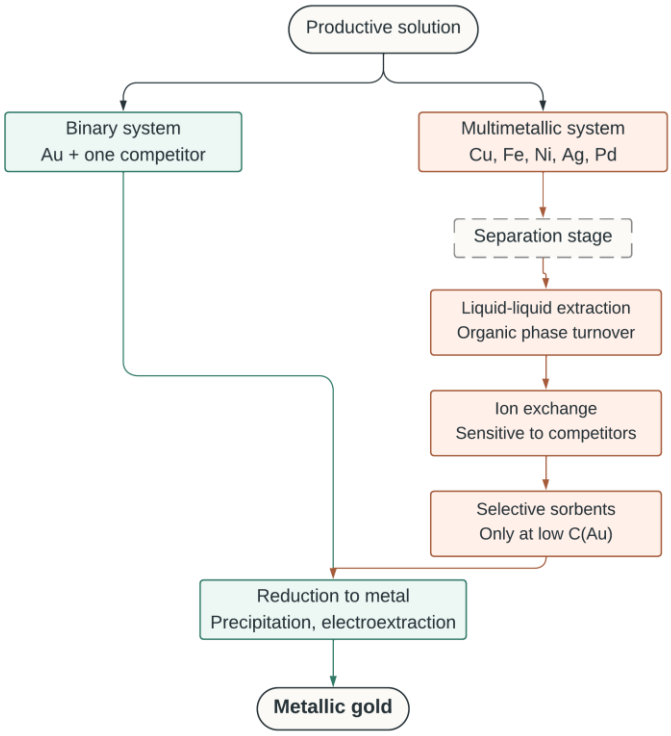

Fig. 5 Selection of the gold recovery route from pregnant solution: in a binary system (gold and a single competitor) the solution passes directly to reduction, whereas a multimetallic system requires a separation stage ***(liquid-liquid extraction, ion exchange or selective sorbents)*** before reduction of gold to the metal by precipitation or electrowinning.

Functionalized resins and porous polymers with a high affinity for gold ions provide selective uptake, in some cases approaching complete recovery, although more often at low gold concentrations and with additional stages for the removal of co-adsorbed ions, which limits their applicability to concentrated industrial effluents [69, 70, 71]. Ion exchange offers good selectivity and is compatible with continuous operation but is sensitive to competing metals [72].

CONCLUSION

Mild flowsheets that do not transform the matrix (glycine, thiocyanate and thiosulfate leaching, together with enhanced-oxidation systems) are attractive for their low reagent toxicity and their operation at atmospheric pressure and moderate temperature. The glycine and thiosulfate systems also tolerate preg-robbing carbonaceous matter and copper. Their limitation is that replacing the ligand does not remove the matrix barrier: gold encapsulated in sulfides or in the refractory phases of anode slime remains inaccessible, and the kinetics are inferior to those of cyanidation. The instability of the reagent itself, most pronounced for thiourea and thiosulfate, increases consumption and complicates process control.

Flowsheets that combine prior matrix transformation with subsequent leaching are more effective: roasting, pressure oxidation or biooxidation for sulfidic primary feeds and alkali fusion for copper anode slime. Decomposition of the sulfide or tin-bearing phase exposes gold and, in certain regimes, allows recoveries above 95% after cyanidation. The advantages of these flowsheets are predictable gold liberation and controllable process parameters. The disadvantages are the high capital and energy costs of the activation stage, an increased impurity load in solution and the need to stabilize arsenic and sulfur.

Integrated pyro-hydrometallurgical chains for electronic scrap (smelting with gold collection in the copper-bearing phase, electrorefining and subsequent anode slime processing) are embedded in existing base metal infrastructure and remove the organic fraction at the pyrometallurgical stage. Their weaknesses are irrecoverable gold losses during mechanical preparation, the number of stages involved and low adaptability to feeds of variable composition.

All routes converge on the same constraint. In multi-metal solutions, separation selectivity limits the outcome. Reduction of gold by precipitation or electrowinning gives more than 99%, and the final result is set by the preceding separation from copper, iron, silver and platinum group metals. Four directions merit further work: reducing the temperature and duration of the activation stage without loss of recovery, closing reagent cycles, increasing the selectivity of sorbents and extractants at industrial gold concentrations, and designing flowsheets around feed variability.

Because the present work is a review, the routes discussed here have not been compared under a single set of experimental conditions, and this defines the agenda for subsequent research. Future studies should benchmark the leaching systems against a common set of criteria: gold dissolution kinetics, selectivity against copper, silver and iron, reagent consumption, environmental impact, technology readiness level (TRL) and economic performance. A comparison of this kind, carried out on identical feed materials, would convert the qualitative picture presented above into a quantitative basis for selecting a flowsheet for a given raw material.

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