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REVIEW PAPER

Current progress on gold recovery from refractory ore and waste electrical and electronic equipment

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Abstract−The physical and chemical properties of gold promote its application, such as in the high-tech, electronic products, and aerospace industries. The easily leachable ore is gradually depleted. Thus, it becomes necessary to extract gold from other resources such as refractory ore and electrical and electronic equipment. The normal method of leaching for gold is cyanide leaching, but it is very dangerous for both environment and operator. Non-cyanide leaching methods, including thiourea leaching, halide leaching, and sulfate leaching have been developed to substitute cyanide leaching. A variety of methods to enrich gold from leaching solutions are described in this paper, including solvent extraction, electrowinning, activated carbon adsorption, and ion exchange resins. Among those methods, ion exchange resins can adsorb gold with high adsorption efficiency and regenerate easily as well. This paper focuses on the research progress of the recovery of gold from non-cyanide leachates by ion exchange resins, summarizes the existing resin types and elution processes, points out the limitations in the application of current ion exchange resins, and discusses possible solutions.

Keywords: Gold, Waste Electrical and Electronic Equipment, Hydrometallurgy, Recovery, Ion Exchange Resin

INTRODUCTION

As an important precious metal (PRM), gold is widely applied in industry, especially in electronic products, because of its unique physical and chemical properties such as high corrosion resistance, stability, good conductivity, and thermal conductivity [1-3]. It is mainly extracted from ore or secondary metal resources by hydrometallurgy. With the easily leachable ore gradually depleting, it is necessary to extract gold from refractory ore and secondary sources [4-6].

Refractory ore is gold ore whose leaching rate is less than 80% by conventional cyanide leaching after fine grinding. It usually has complex impurity metal species. The gold is wrapped with ore and cannot be exposed to a leaching solution by conventional grinding. It mainly includes sulfide ore, carbonized ore, arsenical ore, etc. Secondary sources are derived from urban waste. A large amount of urban waste are produced during urbanization [7], including waste electrical and electronic equipment (WEEE), waste automotive catalytic converters, and some industrial wastes [8]. Electronic products such as cell phones and personal computers contain significant amounts of PRMs such as Au, Ag, Pt, and Pd [9]. According to the statistics, the content of gold in main metal resources such as ore/concentrate is 1-10 g/T, while the content of gold in e-waste is 250 g/T [10]. The value distribution of different electronic waste samples shows that over 70% of the value of mobile phone, calculator, and printed circuit board waste and about 40% of the value of TV board and DVD player [11,12] is come from PRMs, and the content of PRMs in e-waste is comparable to the content of PRMs in ores and concentrates [13,14]. Rational utilization of refractory ore and secondary resources by recovering valuable metal materials can greatly improve the utilization rate of existing resources and realize the sustainable recycling of waste [15].

The enrichment and efficient recovery of gold have attracted the attention of researchers due to limited resources and increasing industrial demand. It is essential to develop simple, efficient, environmentally friendly and highly selective methods for the separation, enrichment and recovery of gold. This review illustrates the progress of hydrometallurgical techniques and compares different extracting methods. Solvent extraction, electrowinning, activated carbon and resin adsorption for gold recovery are illustrated in detail,

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and the recovery method using ion exchange resin is especially focused.

LEACHING OF GOLD FROM REFRACTORY ORE AND WEEE

In the past two decades, hydrometallurgy has become the primary method for recovering gold from ores and e-waste, because it is environmentally friendly and easy to operate [16,17]. The leaching of gold is one of the most important steps in the hydrometallurgical process. Generally, gold leaching methods can be categorized according to the leaching chemicals, like cyanide leaching, halide leaching, thiourea leaching, thiosulfate leaching, etc. The pros and cons of different leaching methods are listed in Table 1.

The selective recovery of metals from WEEE generally involves the conversion of the WEEE from solid to liquid. The common processes are sorting, grinding and/or heating, followed by leaching. Yuksekdage et al. [18] gave the process of physical pretreatment of WEEE (Fig. 1). Firstly, the WEEE was sorted by its composition. Then the sorted materials were crushed and sieved by a plastic grinder. Obtained powder was further ground into finer powder with a vibrating disc mill, and finally the WEEE was sieved through a 500 μm vibrating sieve. At present, metals are mainly leached from crushed e-waste, but the metals will be recovered from whole or bulk e-waste in the foreseeable future. Jadhav et al. [19] used NaOH as pretreatment reagent and removed epoxy solder mask. By treating a 4 cm×4 cm size of PCB with HCl for 22 h, the metals such as gold (Au), platinum (Pd), iron (Fe), nickel (Ni), copper (Cu), which can be completely dissolved.

For the past 100 years, cyanidation has been the main approach to recover gold from concentrates/scrap/ores, which achieves efficient recovery rates through a simple process [20,21]. The reaction formula followed when gold is dissolved in cyanide solution is:

$$4Au + 8NaCN + O_2 + 2H_2O \rightarrow 4NaAu(CN)_2 + 4NaOH \quad (1)$$

It can be known that the gold cyanide leaching process is gold and sodium cyanide reacting to generate soluble and stable gold-cyanide complexes under oxygen. Due to the generation of stable

Table 1. Leaching methods and characteristics

Leaching method	Characteristics	References
Cyanide leaching	1. High selectivity 2. Simple process, high gold recovery rate 3. Toxic 4. Low leaching efficiency for hard-to-leach sulfide minerals containing copper or carbon, etc.	[27,28]
Thiourea leaching	1. Not susceptible to other substances 2. Low toxicity of reagents 3. High reagent consumption 4. Leaching under acidic conditions will corrode the equipment 5. High cost	[29-33]
Halide leaching	1. Fast gold leaching rate 2. Suitable for carbonaceous gold ore, acid-washed gold ore or arsenic-bearing concentrate, etc. 3. Highly adaptable to pH changes 4. Relatively poor selectivity	[34-39]
Thiosulfate leaching	1. Especially suitable for leaching of copper-bearing gold ores 2. Reagents are non-toxic 3. Fast gold leaching speed 4. Non-corrosive to equipment and insensitive to magazines 5. Thiosulfate is unstable and technically demanding	[40,41]

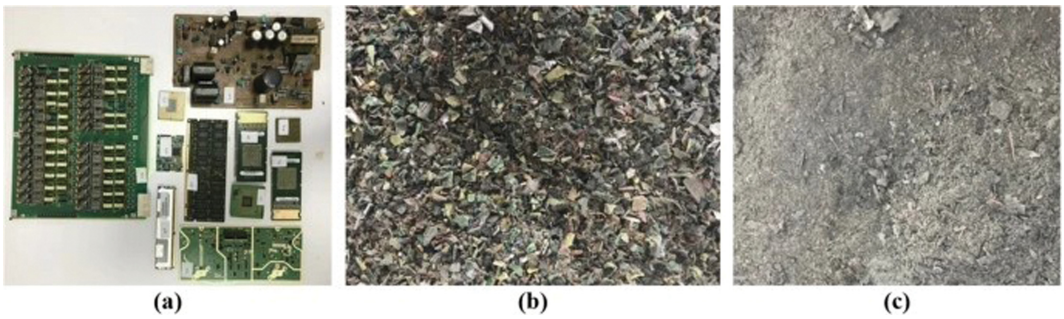

Fig. 1. Appearances of raw (a), crushed (b), and ground (c) e-wastes [18].

complexes, the potential of gold is greatly reduced, so that gold in cyanide solution is easy to be oxidized to $Au(CN)_2$ ligand ion form. Conversion of gold to cyanide gold is basically an electrochemical corrosion process. The interaction of gold and oxygenated cyanide solution is a typical multi-phase reaction carried out in the solid-liquid phase interface because of the existence of crystal defects in the gold particles or surface. The dissolution rate of gold only increases with the increase of cyanide concentration when free cyanide concentration is low. When the cyanide concentration is very high, the dissolution rate of gold depends on the concentration of oxygen in the solution.

In the past 100 years, most of gold has been recovered from sul-

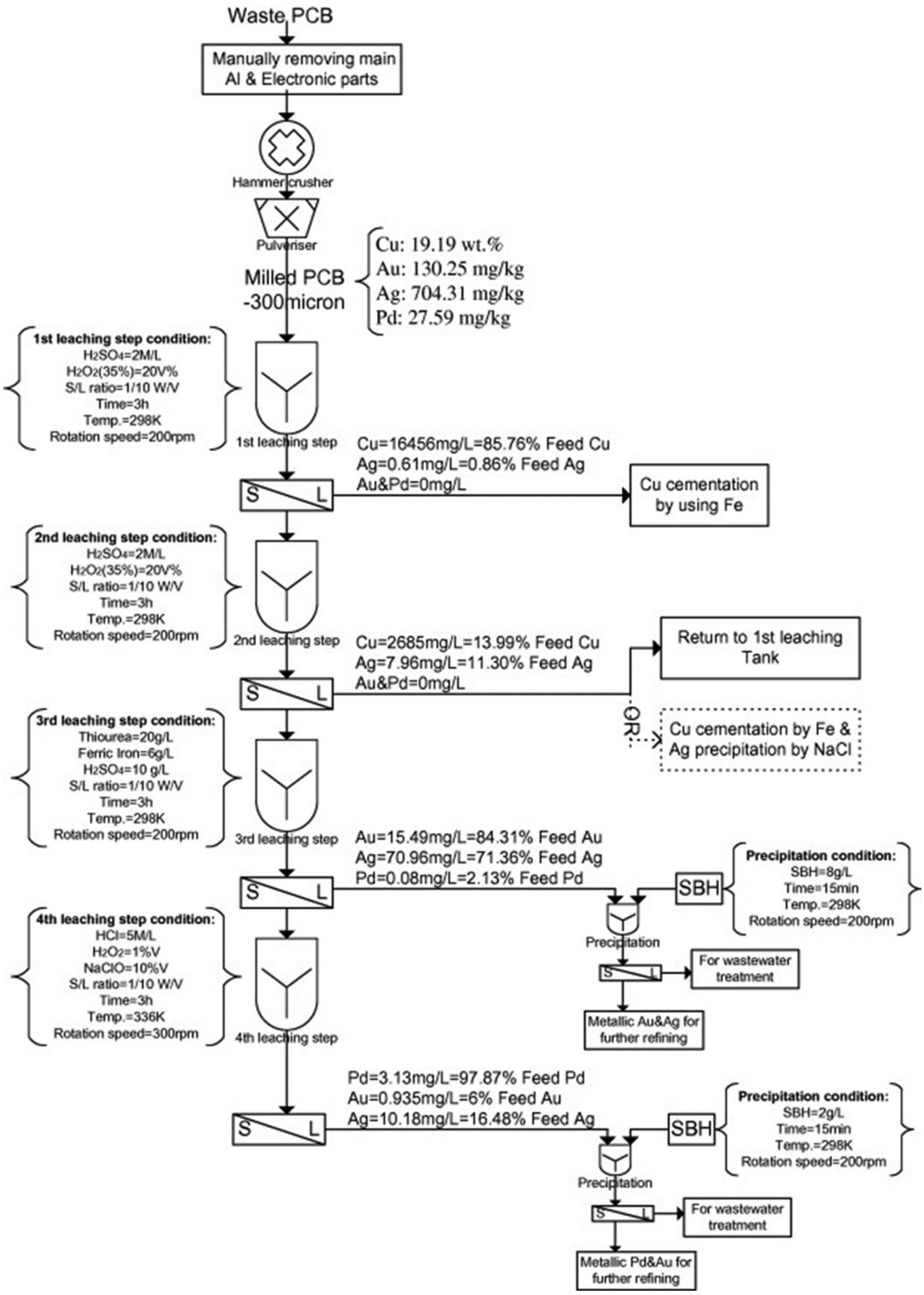

Fig. 2. Process for the recovery of copper, silver, gold and palladium from used PCBs [49].

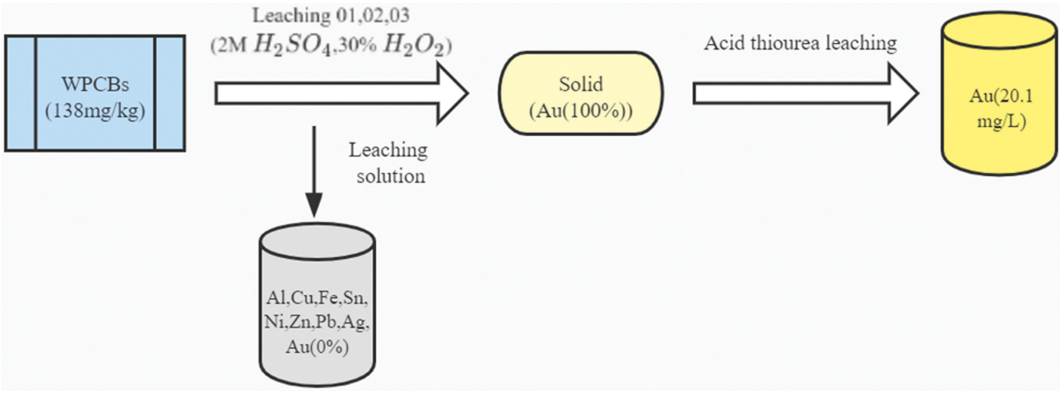

Fig. 3. Three-step cross thiourea leaching.

fide ores, such as pyrite or arsenopyrite. There will always be some sulfur in the leach solution because sulfide minerals have a certain solubility in cyanide solution. Low leaching efficiencies for insoluble sulfide minerals or copper-containing gold resources or carbon pre-leaching are a major disadvantage of cyanide [22]. Jeffrey et al. [22] found that soluble sulfides in sulfide ores can form protective layers of the Au/Sx type to impede the cyanidation leaching of gold. Tan et al. [23] reported that compared with sulfide coating, carbonaceous coating had a greater adverse effect on the dissolution of gold. The results showed that carbonaceous substances may cover the surface of minerals and gold during grinding and mixing, and affect the leaching of gold. It was found that galena could accelerate the dissolution of gold and reduce the instability effect of other sulfides (pyrite, chalcopyrite and sphalerite) that would precipitate as lead sulfide. For example, in the cyanide leaching process of pyrite, the gold extraction increased from 2% to 58% after 10 minutes because of the presence of galena. A study reported different leaching parameters for synthetic seafoam, including dissolved oxygen, pH value, and the existence of lead nitrate and sulfide based minerals regarding the cyanidation process. Gold extraction reached 100% under the following conditions: 500 ppm NaCN, 8 ppm O_2, 500 g/t $Pb(NO_3)_2$, 600 ppm CaO, and no added sulfide components. This study indicated that the presence of lead nitrate, large amounts of dissolved oxygen, and a high pH value (that means high lime concentration) could achieve the maximum extraction of gold [24]. To improve the leaching efficiency, pretreatment is carried out to remove impurity elements from the ore and reduce the effect and consumption of cyanide by biological oxidation process [25], roasting oxidation method and pressure oxidation [26] method before cyanidation.

As public concern and environmental pressure increases [42], researchers along with industry begin to search for other possible leaching reagents. Non-cyanide leaching technology has received a lot of attention [43,44]. Compared with cyanide, many non-cyanide leaching reagents offer a safe and environmentally friendly process with fast leaching kinetics [37,45,46].

Thiourea (also known as thiourea sulfide) has been studied in recent years as a reagent for gold extraction [47]. Ubaldini et al. [48] discovered that the efficiency of gold leaching with thiourea was 87% after leaching at a leaching temperature of 60 °C, 2 hours and pH 1. This study proved that the recovery of gold from low-grade gold-bearing ores is feasible. Gurung et al. [27] recovered PRMs from mobile scrap printed circuit boards (PCBs) by acid thiourea leaching and found that the addition of 0.01 M iron ions to the thiourea leaching solution reduced the complete leaching time from 6 h to 2 h and improved the dissolution rate. Behnamfard et al. [49] selectively recovered copper, silver, palladium and gold from PCBs waste by four consecutive leaching steps (Fig. 2). Birloaga et al. [50] studied oxidative leaching, acid thiourea leaching and thiourea leaching steps for WPCBs. Three stages leaching removed 99% of copper from the solid WPCBs and retained all of the gold (Fig. 3). To further improve the recovery efficiency of gold by thiourea leaching, some researchers pretreated the solution with microwaves [27]. The results showed that 95.5% of gold was leached within 50 min and reached 100% within 70 min, while only 78.1% was recovered within 60 min from the non-microwave treated sample. Qin et al. [51] recovered 98.06% of Au based on the enhancing chlorination roasting process from pyrite. The optimal reaction conditions were as follows: the concentration of thiourea was 7 g/L, solution pH value was 1.2-1.5, the concentration of Fe^{3+} was 4 g/L, S/L ratio was 1/4 g/mL and stirring rate was 300 rpm for 5 h, enhanced chlorination roasting of pyrite with 2 g $CaCl_2$ and 1 g pyrite for 60 min at 850 °C with an air flow rate of 100 mL/min. Lee et al. [52] carried out a two-steps leaching process to deal with WEEE, and the first step was to use 2 M H_2SO_4 and 60 °C for 6 h, the second step was to use 60 g/L thioureas, 2 M H_2SO_4 and 0.5 M H_2O_2 at 60 °C for 4 h, which achieved 95-98% of gold leaching rate.

The key limitation to the application of thiourea leaching is the high price and high consumption of thiourea. To solve this problem, Boboev et al. [53] presented a constructive method for gold recovery from the dump. Leaching rate was 89% and the use of thiourea was declined to only 0.8 kg/t by 0.5% sulfuric acid pretreating the sample before leaching with thiourea and synchronously adding 7 kg/t of ferric sulfate, 12 kg/t of sodium sulfite during the leaching process.

Depending on the chemical conditions of the solution, gold can form gold (I) and gold (III) complexes with iodide, bromide, and chloride [45,54]. Several common halogens have been used in gold extraction. The dissolution reaction of gold is similar for all halogens (Eq. (2)). Birich et al. [55] used bromine for leaching and found that the dissolution rate of gold at 45 °C was 298.05 mg $h^{-1}cm^{-2}$, while other reagents (154.042 mg h^{-1} cm^{-2}) were much lower. Jamie

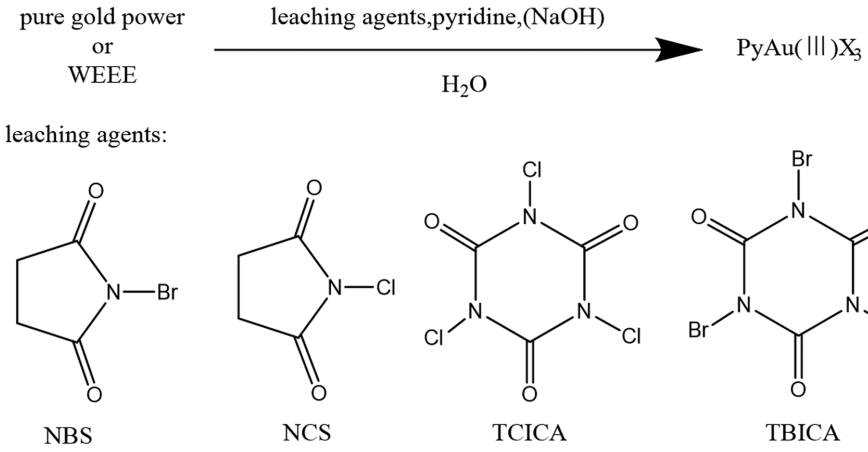

Fig. 4. The structure of leaching agents.

et al. [44] assessed four different organic halides, namely *N*-bromosuccinimide (NBS), *N*-chlorosuccinimide (NCS), trichloroisocyanuric acid (TCICA) and tribromoisocyanuric acid (TBICA) (Fig. 4), as leaching agents for gold recovery from WEEE. The highest leaching efficiency was achieved with NBS, which reached 99% in pure gold powder and 61% in WEEE dust. Whitehead et al. [56] studied the leaching effect of halides on ionic liquid systems. The results indicated that BmimCl/NaI had great potential for effective gold recovery, and the gold extraction rate was 85%. It was recovered from the ore after addition of NaI into BmimCl. The possible reason for the good stability of gold was good, iodide compounds in the ionic liquid. He et al. [57] investigated the chlorination process as an efficient and less polluting method for selective leaching of gold from WEEE. The leaching rate of gold could reach more than 99% after 10 min of pretreating by ball milling at a leaching temperature of 40 °C for 90 minutes, with an initial sulfuric acid concentration of 100 g L^{-1}.

$$Au + 4Br^- = AuBr_4^- + 3e^- \quad (2)$$

The halogens of bromine and chlorine are very fast for gold dissolving, while they are more corrosive. Iodine is the most promising halogen for gold leaching due to its low volatility and low hazard. The dissolution reaction takes place in a neutral or weakly basic solution. Wang et al. [58] studied the application of iodine leaching to gold leaching. With the optimized conditions to treat the actual flotation gold concentrate, the gold leaching rate could reach 85%, and the reaction process of iodine/iodide leaching was shown in Fig. 5.

Thiosulfate leaching is an effective and less harmful process for gold recovery [46,59]. The price of thiosulfate is much lower than that of cyanide, which makes this process very economical compared to the cyanide process [60]. Copper ions are an inhibitor in cyanide leaching of gold, while copper ions can catalyze the dissolution of gold in sulfate ammonia solution [31,40]. Ellen et al. [60] found that providing sufficient oxygen as an oxidant promoted dissolution of gold, but also caused excessive sulfate degradation in the presence of copper ions. Xu et al. [61] investigated the effects of the Cu-NH_3-$S_2O_3^{2-}$ system and Co-NH_3-$S_2O_3^{2-}$ leaching system on gold leaching and sulfate consumption. The use of the Co-NH_3-$S_2O_3^{2-}$ leaching system eliminates the interference of Cu-NH_3-$S_2O_3^{2-}$ to $Au(S_2O_3)_2^{3-}$ recovery, which is expected to solve the problem of high cost and difficult recovery. Furthermore, the electrochemical behavior in the presence of $Co(NH_3)_6^{3+}$ was also investigated in detail. In the presence of cobalt, the coulombic efficiency of gold dissolution (Q_{Au}/Q_T) was much higher than that of Cu(II). At a potential of 250 mV, Q_{Au}/Q_T was about 100% in the initial electrolysis of 300 s catalyzed by $Co(NH_3)_6^{3+}$. The catalytic mechanism for gold leaching is likely to be catalyzed by dissolved oxygen from ammonium thiosulfate. The general reaction equation for the redox cycle between $Co(NH_3)_6^{3+}$ and $Co(NH_3)_x(S_2O_3)^0$ (x=4, 5) is as the following (Eq. (3)-(7)) [62]. Andrea et al. [63] presented a new methodology to recover gold from WEEE using ammonium persulfate ($(NH_4)_2S_2O_8$). To achieve maximum gold recovery, the optimal conditions were simulated using numerical optimization based on the response surface methodology. The predicted value of gold recovery was 100% under a lower concentration of $(NH_4)_2S_2O_8$ (0.65 M), oxygen (0.58 L/min), and L/S ratio (24 mL/g).

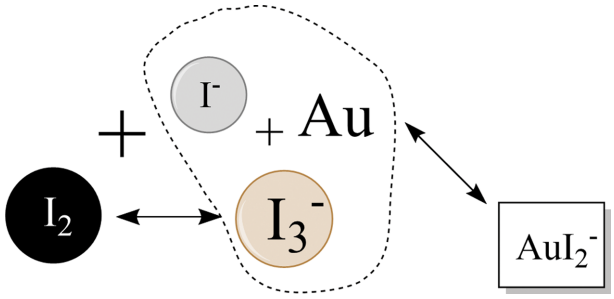

Fig. 5. The reaction process of iodine/iodide leaching.

$$Au + Co(NH_3)_6^{3-} + 3S_2O_3^{2-} = Au(S_2O_3)_2^{3-} + Co(NH_3)_x(S_2O_3)^0 + (6-x)NH_3 \ (x=4, 5) \quad (3)$$

$$Co(NH_3)_x(S_2O_3)^0 + \frac{1}{4}O_2 + \frac{1}{2}H_2O = Co(NH_3)_x(S_2O_3)^+ + OH^- \ (x=4, 5) \quad (4)$$

$$Co(NH_3)_x(S_2O_3)^+ + (6-x)NH_3 = Co(NH_3)_6^{3+} + S_2O_3^{2-} \ (x=4, 5) \quad (5)$$

$$Co(NH_3)_x(S_2O_3)^0 + \frac{1}{4}O_2 + \frac{1}{2}H_2O + (6-x)NH_3 = Co(NH_3)_6^{3+} + S_2O_3^{2-} + OH^- \ (x=4, 5) \quad (6)$$

$$Au + 2S_2O_3^{2-} + \frac{1}{4}O_2 + \frac{1}{2}H_2O = Au(S_2O_3)_2^{3-} + OH^- \quad (7)$$

Different leaching methods have been compared and cyanide leaching methods are being replaced by non-cyanide leaching methods due to their high toxicity to the operator and the environment. The non-cyanidation method has the characteristics of being environmentally friendly, how to recover gold from non-cyanidation leach solution efficiently is the current focus of interest.

CONCENTRATION AND SEPARATION OF GOLD

The methods commonly used to recover gold from leach solution include solvent extraction, electrophoresis, activated carbon adsorption and ion exchange [64-68].

1. Solvent Extraction

Extraction is a method for separating substances from immiscible solution. The extractant combines with metal ions through complexation, association, chelation and other ways, while other metals remain in the aqueous phase and then separate the organic phase to peel gold from it [69]. Solvent extraction has good selectivity and mild reaction conditions. However, the stripping reagent needs a stronger tendency to compound with Au ions, which leads to high costs [70].

Li et al. [71] investigated the methods to extract and strip gold with marketed tertiary amines (Alamine 336, Alamine 308 and Alamine 304-1). It was found that when the molar ratio of Alamine 336/gold exceeded 10, the extraction rate of gold reached 98%, and the acidic thiourea solution could strip gold about 100%. Raiguel et al. [72] compared the extraction efficiency of DEC, methyl isobutyl ketone (MIBK), and dibutyl carbinol (DBC) for gold (Fig. 6). A comprehensive comparison showed that it was greener using DEC as the solvent due to its non-fossil fuel attribute and the gold can be stripped simply with pure water. To recover gold from waste printed circuit board (WPCB) of mobile phone, Mudia et al. [73] used a two-stage acid leaching process to dissolve gold and other metals. It realized the selective separation of gold from leach liquor by solvent extraction using 0.1 M tertiary amide dissolved in toluene. Doidge et al. [74] prepared and evaluated the effectiveness of simple secondary (L^2) and tertiary analogs (L^3) of primary amide L^1 (Fig. 7) for the selective transport of gold in solvent extraction experiments (Fig. 8). The comparison between single and competitive experiments revealed the existence of the third phase. In the single metal solution, the effect of extracted gold was $3^o > 2^o > 1^o$, and due to the formation of the third phase, gold was not transferred from the aqueous phase to the organic phase in the mixed metal solution. The presence of the third phase caused the loss of metals and also illustrated the necessity of solvent extraction experiments in real complex leaching solutions. Kubota et al. [75] extracted Au ions from the diluted aqua regia leaching solution of e-waste by solvent extraction with the newly synthesized extractant N- [N, N-

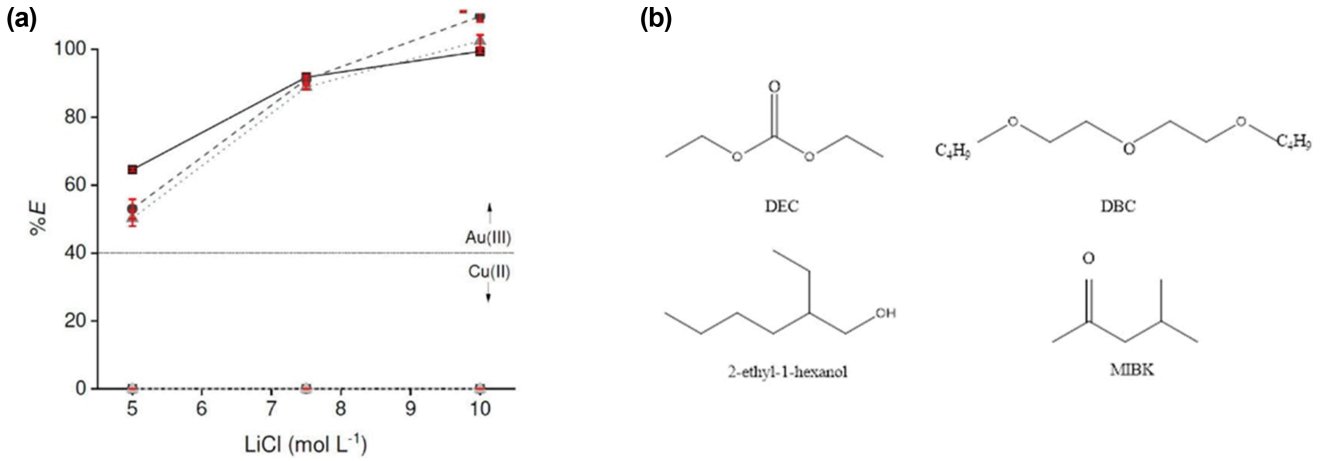

Fig. 6. (a) Percentage extraction (% E) of Au (III) and Cu (II) from LiCl solution with pure DCE (Aqueous composition: 0.1 g L^{-1} Au (III), 0.1 (solid) g L^{-1} Cu (II) (1 (dashed) g L^{-1} and 10 g L^{-1} (dotted)), (b) Structural formulae of DEC, DBC, 2-ethyl-1-hexanol, and MIBK [72].

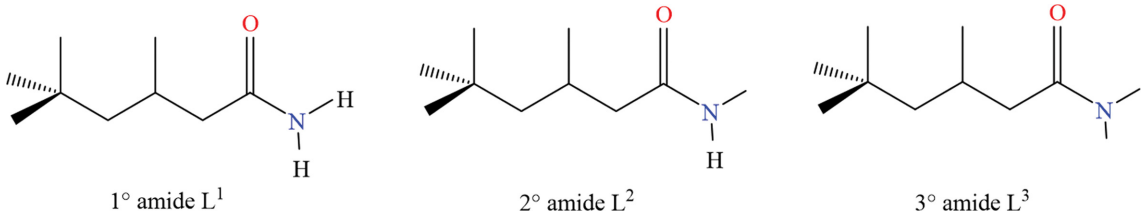

Fig. 7. Structures of 1° amide L^1, 2° amide L^2, and 3° amide L^3 [74].

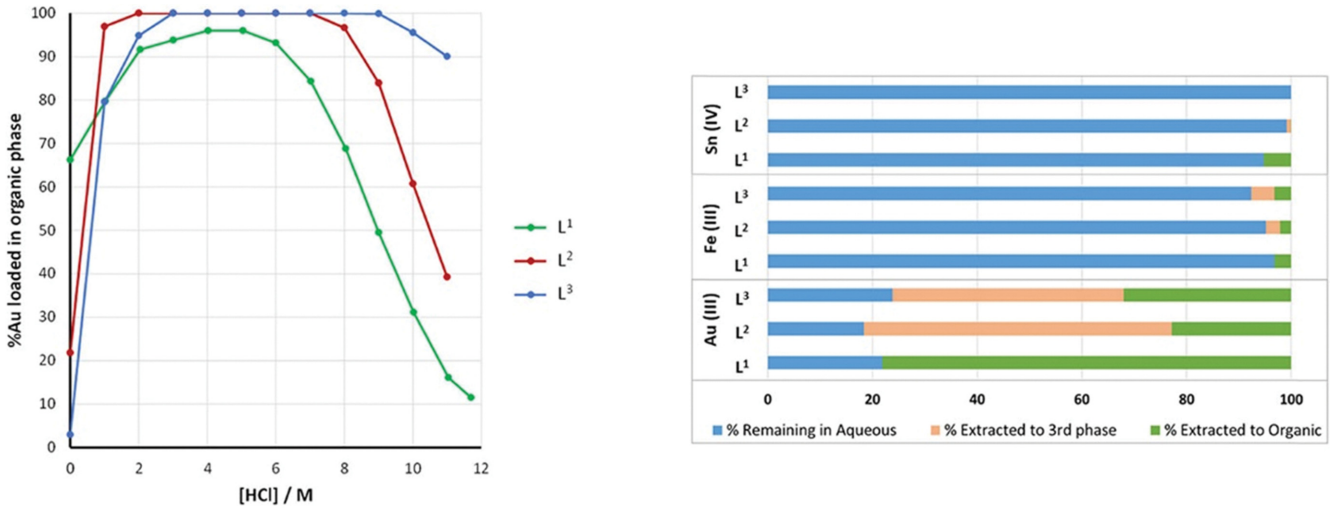

Fig. 8. Effectiveness of extraction in single and complex solutions [74].

di (2-Ethylhexyl) aminocarbonylmethyl] glycine (D2EHAG). It was found that its selectivity was higher than that of a similar solvent extraction system.

However, solvent extraction requires a high concentration of Au ions in the leachate, equipment, and operating costs. Most studies on extractants for non-cyanide leaching solutions are still limited to a single gold solution or a simulated composite solution; application in actual leaching solution needs to be further studied.

2. Electrowinning

Electrowinning is an electrochemical process in which ions form metal deposits by applying different potentials or current intensities [76], which has the strengths of being fast [77,78], simple [79] and low cost [80], with good selectivity [19,81], high product purity [82] and easy to regenerate by a simple polishing of the electrode surface [83].

Steyn et al. [84] investigated the effect of copper on gold electrowinning. In the process of electrolysis, copper will co-deposit with gold (The concentration of metal ions in Eqs. (8)-(11) is 10^{-4} mol dm^{-3}, the concentration of NaCN was 0.2%, and the concentration of NaOH was 2%), which will affect the grade and recovery efficiency of gold, while this influence can be reduced by controlling the cathode potential. Brandon et al. [85] used a cylindrical electrolytic cell with a three-dimensional steel velvet cathode to extract gold from cyanide leaching solution. With the decrease of solution conductivity, more than 96% of Au was recovered from 5 mg/L gold solution in 1 hour under the condition of the current efficiency of 0.33%. Soleymani et al. [86] found that in open circuit potential (OCP) the addition of extra thiosulfate, sulfite, and their mixture shifted OCP graphs from −0.01 V (sole gold-thiosulfate complex) to −0.21 V, −0.33 V, and −0.42 V, respectively. $Au(S_2O_3)(SO_3)_2^{5-}$ was the most stable formation of gold complex. Taskin et al. [87] studied the technological conditions of electrolytic precipitation of gold from thiourea leaching solution. Under the optimal conditions with a total cathode current density of 1,000 A/m^2 and an electrode voltage of 6 V, a gold recovery of 93% could be obtained. Ippolito et al. [88] recovered 95% of gold from WEEE leaching solution after 1.5 h of electrowinning with the power consumption of 3.02 kWh/.

$$Au(CN)_2^- + e^- \leftrightarrow Au + 2CN^- \quad (8)$$
$$E = -0.672\ V_{SHE}$$

$$O_2 + 2H_2O + 4e^- \leftrightarrow 4OH^- \quad (9)$$
$$E = 0.0419\ V_{SHE}$$

$$2H_2O + 2e^- \leftrightarrow H_2 + 2OH^- \quad (10)$$
$$E = -0.809\ V_{SHE}$$

$$Cu(CN)_3^{2-} + e^- \leftrightarrow Cu + 3CN^- \quad (11)$$
$$E = -0.760\ V_{SHE}$$

According to the research on the failure of the gold electrodeposition process, Mohammadi et al. [89] found that the acidification of local solution in pores and crevices would lead to a high corrosion rate of anode made by stainless steel, and the coexistence of gold and typical stainless steel corrosion products led to the loss of gold. The selection of electrode material is the focus of current research. Zeolite is considered to be a very potential electrode material due to its high chemical, thermal and mechanical stabilities. The use of zeolite modified electrodes or surfactant modified zeolite electrodes to improve gold recovery is of critical significance [77,90-92].

3. Activated Carbon Adsorption

Activated carbon is a porous carbonaceous material that can be made from coconut shells with relatively mild adsorption conditions and strong adsorption properties for AuSCN complexes, so activated carbon is generally used to recover gold from cyanide leachates [93]. With the development of cyanide-free leaching technology, the application of activated carbon in other leaching solutions has also been developed [94].

Wang et al. [95] prepared modified activated carbon (SCN-AC) for gold recovery from a thiosulfate solution. It was found that SCN-AC was more suitable for the Cu^{2+}-en-$Na_2S_2O_3$ system when ethylenediamine monohydrate (en) was used as a catalyst instead of NH_3 [95]. Tauetsil et al. [69] used activated carbon to remove gold from aqueous cyanide-glycine solutions, and the gold adsorption followed the Freundlich isotherm model. At a pH of 11, the equi-

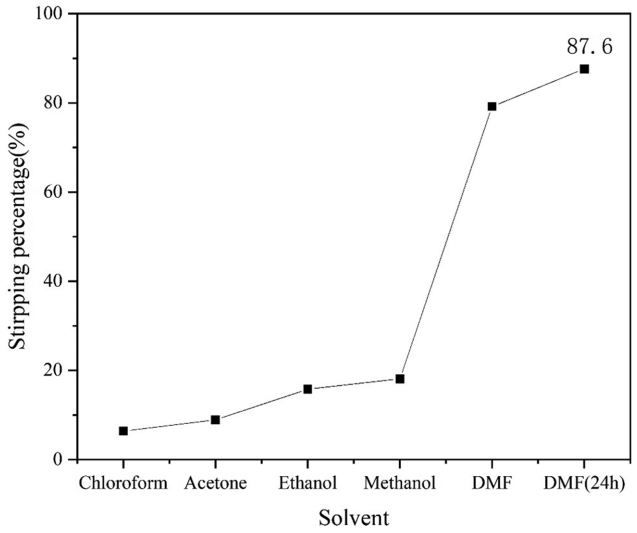

Fig. 9. The results of gold stripping using different solvents at an elution time of 12 h (and DMF 24 h).

librium gold loading capacity (24 h) of the activated carbon was 9.95 kg Au/ton of carbon. Chen et al. [70] studied a clean modification method for activated carbon. Gold was extracted from a thiosulfate system using a synthetic nitrogen sulfur-rich activated carbon and a total gold loading was 25.85 kg/t. Using different reagents to desorb gold, it was found that the ability was positively correlated with the polarity of the solvent. DMF>methanol>ethanol>acetone>chloroform, and the maximum separation efficiency was 87.6% (Fig. 9). Teirlick et al. studied the application of activated carbon in gold iodide solution and the equilibrium loading on the charcoal clearly showed a higher loading profile than gold cyanide [96].

For decades, capturing the dicyandiamide complex $[Au(CN)_2]$ by activated carbon has been the main method for gold recovery. Thiosulfate leaching is currently the most likely replacement for cyanide leaching; however, current commercial activated carbon cannot effectively adsorb thiosulfate gold complexes [97]. The low selectivity of activated carbon is a bottleneck and the high stripping temperature is another [98,99].

4. Ion Exchange Resin

Of the various methods for removing heavy metal ions from solutions, ion exchange is a promising method. The ion exchange method is effective even at very low concentrations of metal ions [100]. Zeolites and ion exchange resins are good ion exchange carriers, with good solid morphology, porous structure, high structural stability, and large specific surface area, being widely used in various industrial processes [80]. Due to the small pore size of zeolites, some substances cannot fully penetrate into the pore channel, which limits its application [101-103]. Ion exchange resin is a kind of polymer with a network structure and functional groups, which can recover gold from a complex mixture system by the reversible ion exchange reaction between the counterions of the resin and the gold complex in the solution [104-107].

Ion exchange resin can achieve high adsorption rates and selectivity by grafting the functional groups which have strong binding forces with ions waiting for adsorption onto the resins. Various resins with amine, sulfur, and aminothiol functional groups were synthesized by cross-linking glycidyl methacrylate with divinylbenzene. The obtained resins were used for the recovery of gold (III) from an acidic solution. The results showed that the resin containing ami-

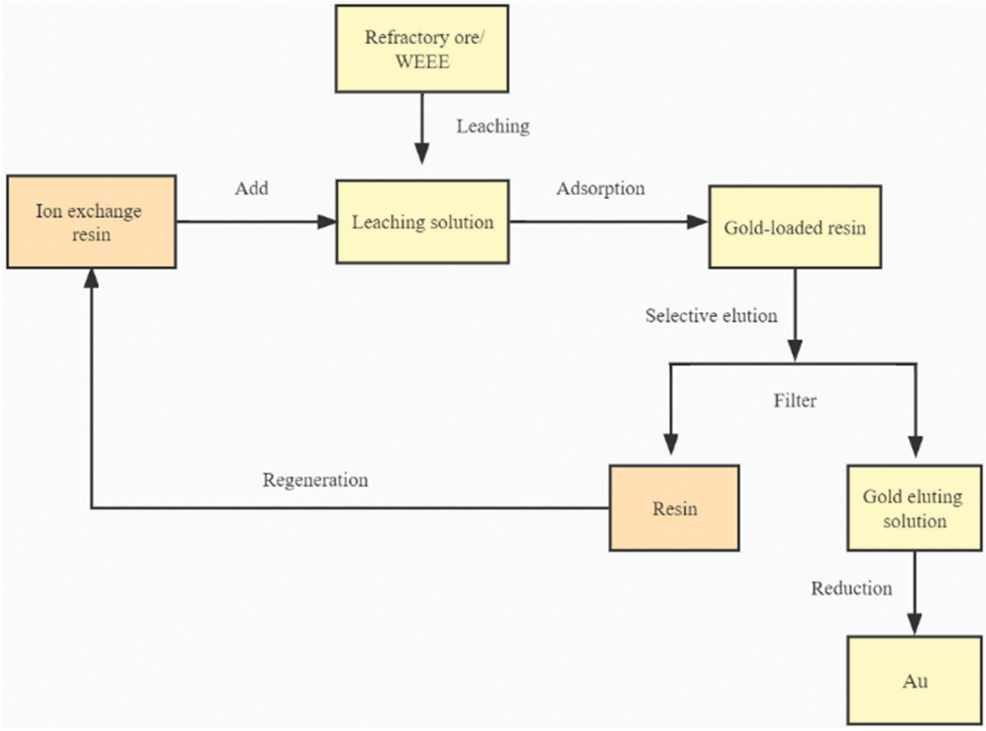

Fig. 10. The process of recovering gold by ion exchange resin.

Table 2. Some applications of ion exchange resins for the removal of gold

Resin	Type	Accompanying elements	Ionic form	pH	Adsorption capacity	Adsorption efficiency (%)	Reference
Amberlite XAD-7	-	Cu, Sn, Fe, Pb	$AuCl_4^-$	2.3MHCl	-	99.8%	[112]
Commercial Cl^- form 717	SBAER	Pd, Pt, Ag	$AuCl_4^-$	3	-	≥99.7%	[113]
Purolite A530	SBAER	Ag	$Au(S_2O_3)_2^{3-}$	10	-	90%	[114]
DTGA-XAD-16	-	Ni, Cu, Sn, Fe, Cr	-	1.0-3.0 M HCl	35 mg/g	-	[115]
Lewatit MP-64	MBAER	-	$AuCl_4^-$	0.03 M HCl	30.5 mg/g −237.5 mg/g	95%	[116]
Dowex 21k	SBAER	Cu	$Au(S_2O_3)_2^{3-}$	11	8.87 kg/t	-	[117]
Amberlite 200	SACER	Fe, Cu, Zn	$Au(CS(NH_2)_2)^{2+}$	1.2-3	-	>98%	[118]
Diaion WA21J	PTAER	Fe, Ca, Ag, Au, Mn, Zn	$AuCl_4^-$	-	-	72%	[119]
Amberjet™ 4200	SBAER	-	$Au(CN)^{2-}$	7.5	-	98.7%	[119]
Purolite A500-2788	SBAER	Ag, Cu, Fe	-	-	-	>98.7%	[120]
	SBAER	-	$Au(S_2O_3)_2^{3-}$	8	2,457 mg/g	-	[121]
Bayer AP-247	SBAER	Cu	$Au(CN)^{2-}$	10.0-11.0	21.4 mg Au/mL-resin	-	[122]
Purolite A500	SBAER	Cu	$Au(CN)^{2-}$	10.0-11.0	15.7 mg Au/mL-resin	-	

Note: WBAER: Weakly basic anion exchange resin, SBAER: Strongly basic anion-exchange resin, MBAER: Medium-basic anion exchange resin, SACER: Strongly acidic cation exchange resin, PTAER: Polyamine type anion exchange resin.

nothiol was more efficient in the recovery of gold (III) with a reported absorption value of 2.23 mmol/g [108]. The adsorption of gold (III), platinum (IV), and palladium (II) on glycine modified cross-linked chitosan resin (GMCCR) was investigated. The optimal range of pH values was between 1.0 and 4.0. And the maximum adsorption capacities of 169.98, 122.47 and, 120.39 mg/g were obtained at pH 2.0, respectively [109].

Resin adsorption has become one of the most attractive technologies for gold recovery due to its rapid absorption rate, high adsorption capacity, mild operating temperature, and ease of regeneration characteristics [110,111]. The process of recovering gold by ion exchange resin is shown in Fig. 10, and the frequently used ion exchange resins are listed in Table 2.

4-1. Gold Recovery from Thiourea Leachate

Gold presents in the thiourea leaching solution in the form of $Au[SC(NH_2)_2]^{2+}$ complexes. The gold complexes in the solution can be almost completely removed by using a cationic resin. The leaching solution can be used again for leaching after removing the gold complex, and the loaded gold on the resin is recovered by burning the resin [123].

Mensah-Biney et al. [124] evaluated the loading capacity of a series of cation exchange resins (Dowex G-23, Dowex G-25, Dowex M-33, Ionac C-249, Ionac CFP-110, Amberlite IR 120, Amberlite 252) and activated carbon for $Au[SC(NH_2)_2]^{2+}$ complexes. After 96 hours at 22 °C, the maximum loading capacity ranged from 189 to 243 kg Au/ton for cation exchange resins and 159 to 208 kg Au/ton for carbon.

Several cation exchange resins were used for $Au(CS(NH_2)_2)^{2+}$ adsorption in batch-type adsorption experiments. It was found that strongly acidic cation exchange resins were the best choice for Au adsorption. Especially, Amberlite 200 was effective for the recovery of $Au(CS(NH_2)_2)^{2+}$ and has outstanding selective adsorption. The acid thiourea leach solution was able to adsorb Au completely in the pH range of 1.2-2.0 [118].

Shields et al. [125] recovered gold from a simulated thiosulfate-thiourea leaching solution, 0.7-0.9 g resin was loaded into a 1.4 mL column, pretreated with 1M H_2SO_4 overnight, and then pumped into the leaching solution for 20 h. The adsorption capacity of resin MTS9140 for gold was 41.6 mg/L.

4-2. Gold Recovery from Halide Leachate

Nguyen et al. [126] used Amberlite XAD-7HP polyacrylate ion exchange resin to recover the gold from the flushing wastewater generated in the manufacturing process of electronic products. The wastewater was strongly acidic (8.0 M H_3O^+). It was found that the pH value of the solution had a great effect on the adsorption capacity of the resin. By adding NaOH or CaO to the solution to neutralize the acidic solution and reduce the acidity of the waste liquid, the adsorption capacity increased obviously. It may be due to the

presence of gold in the form of nonabsorbable $HAuCl_4$ under high acidity in the aqueous phase. When the pH value of the solution was 0.63 (0.23 M H_3O^+), the adsorption rate of gold reached 92.25%, and the highest elution efficiency was 96.96% when the ratio of acetone to acid was 9, meanwhile, the concentration of gold was improved 25-folds than that of the initial solution.

Neto et al. [127] proposed a feasible strategy for gold recovery from WPCB. Strong anion exchange resins DOW™ XZ-91419.00 and Purogold™ A194 were used for Gold purification from multi-metallic solutions. Compared with the initial solution, at least 70% of the gold was recovered, and the obtained solution contained 0.73-1.7 mmol/L of gold with a purity above 92%, and the main impurity was Pb.

Kim et al. [128] used Amberlite XAD-7HP resin to recover gold from chloride leachate with a 95% recovery rate. Two-step elution was adopted, first using 0.1 mol/L hydrochloric acid to elute the copper on the resin, and then using HCl in acetone (1 : 9) solution to elute the gold to obtain a gold solution with a purity of 99.9%.

4-3. Gold Recovery from Thiosulfate Leachate

Compared with the weakly alkaline adsorption resin, the strongly alkaline resin has a better adsorption capacity for gold [129]. When strongly alkaline exchange resins were used to recover gold from ammonium thiosulfate solutions, the results showed that the gold loading was high when copper was not present in the solution, while the gold loading would be reduced when copper was present. The main reason was that copper greatly reduced the stability of the ammonium thiosulfate solution that may oxidize the sulfate to form tetrathionate, leading to competitive adsorption occurring and reducing the adsorption sites of the resin [130].

The adsorption performance and elution behavior of the gel-type resins Dowex G51, Dowex 21K and Amberlite IRA-410 towards gold from sulfate solutions containing gold or gold and copper were investigated. In the presence of copper, the tetrasulfate was effectively removed at pH 11, allowing for effective loading and high selectivity for gold. In the absence of copper, gold can be loaded rapidly from the sulfate solution onto the strongly basic ion exchange resin with a high loading concentration. The most effective eluent was sodium sulfite/ammonia combination, but the sodium chloride/ammonium thiosulfate combination was probably the least costly, and the addition of sodium thiosulfate to the eluent appeared to hurt the elution efficiency of gold [117].

Gamez et al. [131] recovered gold from secondary sources of ammonia-sulfuric acid solution using MTA5011 ion exchange resin. The high concentration of copper in the PCB affected the dissolution of gold, so the concentration of copper was controlled to 1% by removing some of the copper with nitric acid, and the recovery of gold was improved at a higher resin/solution ratio. The optimum conditions were resin/solution ratio of 100 g/L, stirring at pH=10.5 and room temperature for 24 hours. Under these conditions, 87% of the gold on the resin could be recovered.

Xu et al. [61] investigated the difference between cobalt-ammonia complex as the oxidant and traditional copper-ammonia catalysis. In contrast to copper, cobalt was difficult to be loaded on the strongly basic anionic resin, which allowed the resin to load gold very quickly, reaching 96.2% in only 20 minutes. Furthermore, the gold can be stripped by a simple one-step elution process due to the low loading of cobalt onto the resin. For the elution of the copper-catalyzed system, a pre-elution of copper was required to separate copper and gold first. In addition, they also developed a nickel-catalyzed ammonium thiosulfate leaching process to recover gold, the loaded gold can be eluted by a simple one-step process because the nickel did not co-adsorb with the gold (Fig. 11) [132].

Jeffrey et al. [121] investigated the elution process for recovering thiosulfate gold from ion exchange resins and developed a new elution technique of sulfite enhanced chloride to improve the elution efficiency of gold by synergistic ion exchange. When the sulfite was added, $Au(S_2O_3)_2^{3-}$ was converted to $Au(S_2O_3)(SO_3)^{3-}$ and the affinity for the strong anion exchange resin was reduced. And the adsorption experiment was carried out for 22 consecutive days, and the generated resin was returned for adsorption after elution to obtain a concentrated eluate, and then the gold was recovered by the electrowinning technique.

Porous metal organic frameworks (MOFs) consist of metal ions/metal clusters and organic ligands with large specific surface area, large porosity, and adjustable structure and function, thus has been widely used in adsorption and separation [133,134], biomedicial [135,136], catalysis [137], and sensing fields [133,138]. Resins are the most important catalysts or carriers to improve the functionality

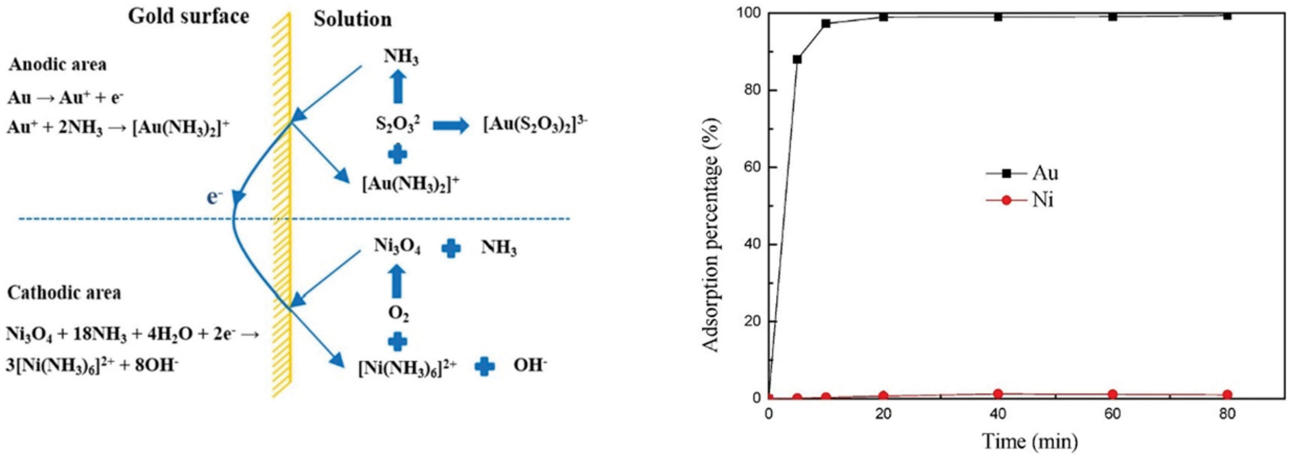

Fig. 11. Mechanistic modeling and resin adsorption effect of gold leaching from sulfate under nickel-ammonia catalysis [132].

of MOFs. There are few reports on the application of gold nanocluster-MOF nanocomposites [139-141], but reducing the development cost and high gold loading of such materials are still a focus of attention. Wu et al. [142] used thiourea-modified MOFs materials to extract Au from WEEE with an adsorption capacity of 326 mg g^{-1} for Au at pH 4. Au NCs/UiO-66-TU was obtained by sodium borohydride ($NaBH_4$, 10 mM) to reduce the PRM ion for 0.5 h.

4-4. Elution of Gold and Regeneration of Resin

The elution process and the regeneration of resin after adsorption are important means to prolong the service life of resin and reduce the use cost. A study showed that the loading capacity of the resin was reduced by 50% after the 14-cycle process, which equates to 3.5% of the total capacity of resin lost per loading/ elution cycle or about one-third of the total cost of one cycle. This shows that minimizing resin consumption is a key issue that must be addressed for resin applications. The influence of adsorbent substances and eluent on the resin during the adsorption/desorption process is one reason for the consumption of resin, the other may be due to the mechanical strength of the resin not being good enough. Under these conditions, the adsorption sites and loading capacity of the resin were reduced.

Therefore, the first and most important thing is to choose a moderate elution method and a more effective eluent to reduce the impact of the elution process on the resin. Table 3 summarizes the elution procedure for desorption of gold from ion exchange resins. As listed, hydrochloric acid and thiourea were the common elution reagents. In addition, the mixture of thiourea and organic solvents, Na_2SO_3, $NaNO_3$ and thiocyanate were also studied as elution reagents. For instance, Zhang et al. [117]studied the adsorption-desorption behavior of gold onto ion exchange resins Dowex 21K and G51. It was found that gold could be loaded quickly from pure solution onto the resin, while the resins had a reduced capacity for gold adsorption when it was simultaneous loading of copper and gold. To study the selective elution process, tetrathiocyanate, which has strong competition with gold for adsorption, was first selected as the eluent. The elution was incomplete at low doses, with total elution rates of 95.3% (G51) and 98.7% (21K) at 55 BV, respectively. The use of thiosulfate was found to be more effective for gold and copper elution, with no obvious tail in the elution curve and a smaller dosage than that of tetrathiocyanate.

On the other hand, improving the mechanical properties of the resin is another direction to strive for and can be further investigated in the following aspects. First, some researchers studied the effects of different polymeric monomers on the mechanical strength of the resin. It was found that styrene-based vinyl esters have better mechanical properties than methyl acrylate as a copolymer monomer [143]. Secondly, the mechanical strength of the resin is improved by adding reinforcing materials. Reddy et al. [144] prepared composites by adding 20 wt% cordia dichotoma fibers and 15 wt% granite powder to polyester resin, which improved the mechanical properties of the composites. Thirdly, the existing resins can be modified to improve its mechanical strength. Arasaretnam et al. [145] modified phenol formaldehyde resole resin using boric acid, sulfuric acid and lignin biomass to improve the mechanical properties of the resins. Boron-modified phenol formaldehyde resins were prepared by using boric acid with the formalin method, and the obtained resins have smooth surfaces and higher mechanical strength.

Table 3. The elution process of gold adsorption using ion exchange resins

Resin	Flow rate	Eluent	Desorption efficiency	Reducing precipitation reagent	Recovery efficiency	Reference
Amberlite XAD-7	3 BEV/h	Acetone-hydrochloric acid	99.5%	5% hydroquinone solution	99%	[112]
Amberlite IRA-35	1 mL/min	Thiourea, Ethyl alcohol	98.7%	-	-	[146]
Lewatit MP-64	-	Acidic thiourea solutions	-	$NaBH_4$	90%	[116]
Dowex 21k	2 BV/h	Na_2SO_3, NH_3	99.9%	-	-	[117]
Amberlite 200	-	$Na_2S_2O_3$	100%	-	-	[118]
Diaion WA21J	-	Thiourea	98%	$NaBH_4$	97.6%	[119]
Purolite A500-2788	10 m^3/h	$Na_2Zn(CN)_4$	80 mg/L	-	-	[120]
	-	Chloride, Sulfite	-	Electrowinning	97.8 wt%	[121]
Bayer AP-247	-	$NaNO_3$	91.0%	-	-	[122]
Purolite A500	-	Thiocyanate	90.0%	-	-	[122]

"-": Unknown.

CONCLUSIONS

Gold is a rare and non-renewable PRM with indispensable applications in many industrial fields. Gold-bearing ores and e-waste often contain other metal ions such as copper, iron, and silver. Therefore, choosing effective leaching and separation methods is of great importance for the utilization of gold.

At present, gold recovery is mainly focused on refractory ores and scrap metal resources, of which two factors must be taken into account. First, a careful understanding of the elements contained in the gold-bearing resource and the availability of the resource must be considered to determine potential leaching options as those outlined in this review. For cobalt- and nickel-bearing resources, the catalytic effect on the leaching process is important. For locations where energy costs are high, ultra-fine grinding may not be a viable option. Second, an appropriate recovery scheme must be selected based on the element type and concentration of the leachate. How to collect elution needs to be considered in particular, as large wastewater discharges are not allowed from an environmental and social point of view. New processes for the application of ion exchange resins in gold recovery need to be explored actively. To improve the adaptability of ion exchange resins to complex gold-containing solutions, the new processes should revolve around how to adsorb gold with high selectivity and reduce the costs of the resins.

At the time of submission of this review, certain key topics have not been adequately addressed in previous publications. For example, the recovery of gold from cyanide leach solutions has been the focus of previous research, leaving important knowledge gaps for the next research. This is particularly important, since it is well known that research has been conducted on cyanide alternatives due to the dangers of cyanide. On the other hand, with the gradual depletion of easily leachable ores, the focus of the leaching process is shifting to other ores and gold-bearing resources, where the influence of other metals, such as iron, zinc, and copper, on the adsorption selectivity and elution of the resin is a challenge for the application of the resin. Therefore, to have a deeper understanding of gold extraction from refractory ores and waste electronics, further research on the above aspects is needed and important.

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AUTHOR STATEMENT

All persons who have made substantial contributions to the work are reported in the manuscript. All authors have approved the final version to be published.

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