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Development of an integrated model to recover precious metals from electronic scrap - A novel strategy for e-waste management

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Abstract

The authors in this paper makes an attempt to propose a novel integrated model for the recovery of precious metals (gold - Au and silver - Ag) for the management of e-waste using a combination of hydrometallurgical (chemical) and biometallurgical (low cost biomass) processes. Chemical process involves leaching of gold/silver from electronic scrap using cyanide. The leachate containing gold/silver complex thus obtained is then subjected to biosorption process using low-cost biomass. The model proposed by us will help to strengthen the 'processing' part of the functional elements employed for proficient e-waste management. Feasibility study was also conducted to explore the possibility of removal/recovery of silver-cyanide using low-cost biosorbents. Results showed that *Eicchornia* root biomass and Waste tea powder were efficient biosorbents for leached silver-cyanide from electronic scrap. The concentrated silver-cyanide recovered could further be used as an input material for electroplating industry.

1. Introduction

Waste Electrical and Electronic Equipment (WEEE) or e-waste, like municipal solid waste (MSW), is one of the fastest growing advanced type of solid waste streams in the urban environment worldwide. E-

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wa0ste is a generic term encompassing various forms of electrical and electronic equipments (EEE) that are old, end-of-life electronic appliances and have ceased to be of any value to their owners (UNEP, 2007). E-waste/WEEE in general comprises of all old, end-of-life appliances like computers, laptops, audio and video products, refrigerators, freezers, mobiles phones, etc. along with their peripherals. The sources of e-waste are relatively expensive and essentially durable products used for data processing, telecommunications, or entertainment in private households and businesses.

Globally, WEEE is growing by about 40 million tons a year (Wath et al., 2010). In developed countries, e-waste constitutes 1-2% of the total solid waste generation. In US, it accounts for 1-3% of the total municipal waste generation. In European Union (EU), total amount of e-waste generation ranges from 5-7 million tons per annum or about 14-15 kg per capita and is expected to grow at the rate of 3-5% per year. Developing countries are also in the challenging phase as they are already facing the continuum of hazardous e-waste mountains (UNEP, 2007). In South Africa and China for example, the report predicts that by 2020, e-waste from old computers will jumped by 200 to 400 percent from 2007 levels and by 500% in India (UNEP, 2007). The growth rate of discarded electronic waste is high in India since it has emerged as an Information Technology giant and due to modernization of lifestyle. The total e-waste generated in India amounts to > 1,46,180 tons per year as on 2010 (Wath et al., 2011).

The physical composition of e-waste is very diverse and contains over 1000 different substances, which falls under organic and inorganic fractions. Heavy metals form a significant part of inorganic fraction accounting for 20-50%. E-waste consists of hazardous metallic elements like lead, cadmium, chromium, mercury, arsenic, selenium and precious metals like silver, gold, copper and platinum. Overview indicates that manufacturing of mobile phones and personal computers consumes 3% of gold and silver mined worldwide each year; 13% of the palladium and 15% of cobalt. Whether hazardous or precious, heavy metals are non-renewable and finite resource and therefore eventually become very valuable. Moreover, managing e-waste is a confounding task due to the various challenges like technical, financial, strategic, information failures, etc. It is, therefore an urgent need to manage e-waste in a formal, systematic and eco-friendly manner by way of removing/recycling the precious metals from waste streams. In emerging economy like India, current practices of e-waste management are followed completely in disorganized manner which may cause deleterious impacts on human health and ecology. It was thought that if an efficient system for removal/recovery could be proposed and developed, precious metals could be conserved, which in authors opinion would be a novel approach of resource recovery.

In the present paper, the authors attempt to propose a novel integrated model for the recovery of precious metals (viz. gold and silver) for the management of e-waste using a combination of hydrometallurgical (using chemicals) and biometallurgical (using low cost biomass) processes. The integrated model proposed by us would help to strengthen the 'processing' part of the functional elements employed for proficient e-waste management. Feasibility study was also carried out to explore the possibility of removal/recovery of silver-cyanide from leachate solution using low-cost biosorbents.

2. Gold and silver leaching from electronic scrap by hydrometallurgical (chemical) process

As mentioned earlier, heavy metals form a significant part of inorganic fraction accounting for 20-50% of the total e-waste being generated. Apart from hazardous metals (viz. lead, cadmium, chromium, mercury, arsenic, selenium) many other economically important metals like gold, silver, copper and platinum are also present in e-waste. Whether hazardous or precious, heavy metals are non-renewable and finite resource and therefore contribute the most value in electronic scrap. And from the economic point of view, recovery of precious metals from e-waste is one of the most attractive options. Significant literature has been cited on the recovery of precious metals using hydrometallurgical techniques and has been one of the most active research areas since 1980s (Macaskie et al., 2007; Ogata & Nakano, 2005).

Hydrometallurgical methods when compared with conventional pyrometallurgical methods are more accurate, predictable, and can be controlled easily (Cui & Zhang, 2008). Main steps in hydrometallurgical processing consist of leaching, purification, precipitation, solvent extraction, adsorption and ion-exchange to isolate and concentrate the metals of interest. The solutions consequently are treated by electro-refining process, chemical reduction, or crystallization for metal recovery (Ritcey, 2006).

The first step in hydrometallurgical processing is the leaching of metals using suitable solvent. The most commonly adopted leaching agents in recovery of precious metals is cyanide. Halide, thiourea and thiosulfate are also used (Sheng & Etsell, 2007). Cyanide is being employed for extraction of gold and silver in mining industry for more than a century and is a low-cost option (Hilson & Monhemius, 2006). The mechanism of gold/silver dissolution in cyanide solution is an electrochemical process. The process involves reaction between gold, oxygen, cyanide and water (Rajeshwari, 2008). The overall reaction is shown below.

$$4Au \ + \ 8CN^- + O_2 + 2H_2O \ \rightarrow \ 4Au(CN)_2^- \ + \ 4OH^- \tag{1}$$
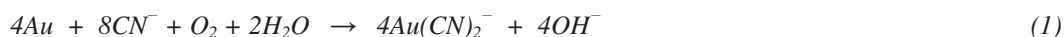
Maximum dissolution of gold, silver and other precious metals in cyanide solution occurs at pH above 10. Cyanide, although a very useful chemical from industrial point of view, also enjoys the higher status due to its evil reputation in the past. Bhopal Gas Tragedy, Bhopal, India (1984); Green Spring Gold Operation, Ely, Nevada, USA (1992); and Szamos-Tisza Cyanide Pollution, Romania and Hungary (2000) are some of the examples, wherein either organic or inorganic cyanides have played the key role. These accidents have killed and affected large number of people, birds, other flora and fauna, severe contamination of surface and groundwater has precipitated the widespread concern over the use of cyanide as a leach reagent (Hilson & Monhemius, 2006). Therefore, many non-cyanide substitutes have been reported of which thiourea and thiosulfate is regarded as being the most realistic substitutes. Several research investigations have also been undertaken to determine the effectiveness of chlorine as non-cyanide substitutes. However, it is difficult to employ it (and therefore not in use) for two main reasons: (i) special steel and rubber lined equipment is required to resist oxidizing conditions and highly corrosive acidic nature; (ii) Involves health risk due to poisonous nature of chlorine gas.

Thiourea, although an effective reagent for gold leaching, very few full-scale operations are in existence and are only restricted to developed nations. Commercial applications of thiourea for gold leaching has been hindered for the following reasons (Cui & Zhang, 2008): (1) thiourea is more expensive than cyanide; (2) since thiourea readily gets oxidized in solution its consumption in gold processing is very high; and (3) steps involved in gold recovery process requires more development. Because of these reasons, the thiourea process is still in its infant stage.

Another non-cyanide substitute chemical widely researched out is thiosulfate ($S_2O_3^{2-}$) (81-97). It has large number of applications in photography units and pharmaceutical industries. Although thiosulfate has the potential environmental benefits it has several disadvantages: (i) high thiosulfate consumption during extraction (ii) the leaching process (reaction rate) is very slow (iii) requires copper (II) as an oxidant, which is again a non-renewable and precious metal resource and, (iv) the process uneconomical. Currently, no simple and affordable method for recovering gold/silver from thiosulfate leach solutions exists. The authors further argues that high consumption of thiourea/thiosulphate during the leaching process would probably consume more energy and other supplementary resources for its formulation and further use during it entire life cycle assessment (LCA) when compared with cyanide.

In the light of above account, use of cyanide has several advantages over various non-cyanide substitutes like thiourea, thiosulphate and chloride ions. These could be classified into primary, secondary and tertiary (ultimate) advantages and has been depicted in Table 1.

Although cyanide has several advantages, using it as a leaching agent has some disadvantages. Cyanide is deadly poisonous chemical and therefore requires careful handling, storage and transportation.

Nevertheless, improper management of cyanide may cause deleterious impact on human health and ecology. Moreover, the low-tenor cyanide effluents emanated by the industry require stringent control and statutory compliance. Inspite of the fact that cyanide is potential dangerous, it could be managed effectively by adopting sound management practices by way of using personal protective equipments (PPEs) at workplace, appropriate containers (avoid metal containers) for storage, proper transportation facilities and cost-effective treatment of cyanide effluents. Considering the above mentioned merits and demerits of cyanide, the authors strongly suggest the use cyanide for the leaching of gold and silver and therefore recommend the same in the proposed integrated model. In a developing and emerging economy like India, waste management has to be a low cost proposition for its wider acceptance and utilisation.

Table 1. Primary, secondary and tertiary merits of use of cyanide for metal leaching in the current scenario

Primary (direct) advantages	Secondary (indirect) advantages	Tertiary (ultimate) advantages
Cyanide is a good complexing agent; forms stable metal-cyanide (M_xCN) complex with precious metals like gold, silver and many other metals	Recovered gold/silver cyanide complex after leaching could be used as a raw/input material for electroplating industry	Will reduce the demand for natural resources significantly
Reaction rate of cyanide with precious metals is moderately rapid with non-cyanide substitutes	Will enhance the profit share/margin of the organisation and economic growth of the country	Will help to conserve the metal resources
Use of cyanide does not require oxidant or catalyst or any other resource other than water	Will strengthen and create a sound industrial network/ecosystem	Protect the environment
Use of cyanide is an effective low-cost solution		

3. Recovery of precious metals by biometallurgical process with special reference to biosorption

   Recovery of metals by 'bio' component has been one of the most promising technologies of the last decade (Macaskie et al., 2007). Biometallurgy has immense potentials for major breakthrough for the materials and minerals processing industries and being researched out from biochemical perspective for the last two decades. And many national and international organisations are showing interest in this new technology. Currently, research and development is in progressive stage for a number of metals such as copper, cadmium, zinc, nickel, cobalt, chromium, gold and silver (Morin et al., 2006).

   There are two main areas of biometallurgy for recovery of metals viz. bioleaching and biosorption. Research studies show that for recovery of gold and silver, the activity of leaching bacteria is applied only to remove interfering metal sulfides from ores bearing the precious metals prior to cyanidation treatment (Morin et al., 2006). Although having huge potentials, bioleaching of metals involves use of live microorganisms (viz. bacteria, fungi and algae), which incurs significant time to complete the process. Moreover, live microorganisms are subject to toxicity of metals and other chemicals indicating that further development in the area of bioleaching is required. It is for this reason; the researchers, in the present paper suggest leaching of gold and silver using cyanide (chemical process) for our proposed model.

   Interactions of biomass with anionic and cationic chemical species (viz. heavy metals; pesticides, etc.) are well known and; they offer numerous opportunities of exploiting the "variety of natural biomass" for environmental clean-up. Biomass (both live and dead) has an array of defence mechanisms, which keep the target chemical ions trapped either outside or inside the cells. It is widely known that biomass of all types (bacteria, fungi, yeasts, algae, plants, organic waste, etc.) have the ability to bind to metals, in some cases selectively, from aqueous solutions (Paknikar *et al.*, 2003). This phenomenon is termed as 'biosorption' and the biomass responsible for the process are known as 'biosorbents'. The biosorption phenomenon is the combination of processes such as electrostatic interactions, ion exchange,

complexation, formation of ionic bonds, precipitation, nucleation, etc. Biomass surfaces are usually charged and the functional groups like phosphoryl, carboxyl, sulphahydryl and hydroxyl of membrane proteins, lipids and many other components are responsible for adsorption of many organic and inorganic chemical species. The overall interactions are a result of complexity of biomass surfaces and chemical / physical properties of the target ions (Modak and Natarajan, 1995).

The phenomenon of biosorption could be exhibited by active (i.e. living) as well as passive (i.e. non-living) biomass. The advantages of biosorption are: (i) non-living biomass are not subject to toxicity limitations; (ii) costly nutrients and aseptic conditions are not required for the cultivation of biomass; (iii) the process is very rapid; (iv) waste from fermentation industry and nature could be cheap source of biomass; (v) biosorbent could be operated at wider range of pH, temperature and target compound concentration; (vi) established theories, conventions and formulae could be applied to the adsorbent (Paknikar *et al.*, 2003).

4. Feasibility study: Biosorption of silver-cyanide from solutions using low-cost waste biomass

For the present study, variety of low-cost biomaterials (Table 2) were used obtained/ collected from diverse source and habitats. Broad categories of the biomaterials used were as follows: (i) Agricultural by-products, (ii) Industrial waste/by-products, (iii) Solid waste, (iv) Waste fungal cultures, (v) Mixed algal biomass, (vi) Photosynthetic plants and, (vii) some adsorbents as reference materials (Gaddi and Patil, 2011). Primary processing of the biomass samples (collection, transportation, washing, drying, grinding) was carried out as per the methodology previously prescribed by Gaddi and Patil (2011). Silver-cyanide [Dicyanoargentate – DCAG i.e. $Ag(CN)_2^-$] solution was prepared stoichiometrically as per the method of Patil and Paknikar (2000). Analytical grade chemicals and borosilicate glassware were used for all the experiments. Prepared stock solutions and reagents were stored in refrigerator (5-10°C). Silver was estimated by Atomic Absorption Spectrophotometer (Elico, India), while the total cyanide content was estimated according to the method prescribed in Standard Methods (APHA-AWWA-WEF, 1998).

Biosorption study on silver-cyanide was conducted by batch equilibration method as prescribed by Gaddi and Patil (2011). The amount of silver-cyanide sorbed at equilibrium, Q (mol/g), which represents the Ag-cyanide uptake, was calculated from the difference in DCAG concentration in the aqueous phase before and after adsorption, according to the following equation:

$$Q = V (C_i - C_f) / 1000 \, m \qquad (2)$$

Where, Q is the DCAG uptake (mM per gram biomass); V is the volume of DCAG solution (ml); C_i is the initial concentration of DCAG (mM); C_f is the final concentration of DCAG (mM); m is the mass of sorbent (g). Table 2 depicts the results obtained for DCAG sorption under optimal pH conditions. The results showed that optimum Q (i.e. μmol DCAG sorbed per gram biomass) of 0.1 mM DCAG for most of the waste biomass/sorbents tested were at pH 6.0. Biosorption was significantly decreased above pH 7.0 and increased in acidic conditions. Other than the reference material (i.e. activated charcoal), which showed highest sorption (5.00 μmol/g), biomass of *Eichornia* roots and Tea powder waste were the most efficient biomaterials for DCAG among all the biomass screened. The Q values obtained for efficient biomass were marginally below the Q value of activated charcoal. The optimum pH for maximum DCAG biosorption by *Eicchornia* root biomass and Tea powder waste biomass was in the range of 6.0 to 9.0 and 5.0 to 9.0, respectively.

It is well documented that the process of biosorption is influenced by the solution pH (Paknikar et al., 2003). The increased sorption of DCAG under acidic conditions may be attributed to the protonation of biosorbent surface (i.e. the functional groups) acquiring net positive charges and therefore, probably the formation of species such as H^+-$AgCN_2^-$ on the biomass might have taken place. Thus, relatively more DCAG species could have accommodated on the biosorbent sites. Most biomass from natural origin

contains large number of surface functional groups (hydroxyl, carbonyl, carboxyl, sulphydryl, amine, imine, amide, phosphodiester, etc). Probably some of these functional groups might have played the key role in the sorption of DCAG. Experimental controls run simultaneously confirmed that biosorption was the only operating mechanism for the removal of DCAG from solutions. Furthermore, DCAG being a stable metal cyanide complex (logK 20.8) as against free cyanide ions (logK 9.2), does not form toxic hydrogen cyanide (HCN) gas under acidic conditions of biosorption process thereby making it a safe procedure to operate. On the basis of maximum DCAG uptake values obtained at optimum pH, *Eicchornia* root and Tea powder waste biomass were selected for further experiment of DCAG loading capacity and the results are shown in Table 3.

Table 2. Biosorption of DCAG at optimum pH

Sr. No.	Biosorbent	Q (µmol MCN sorbed per gram biomass)	Sr. No.	Biosorbent	Q (µmol MCN sorbed per gram biomass)
(A)	Agricultural waste/by-products		16.	*Penicillium* waste	3.99 (6.0)
1.	Coconut fibres	4.62 (5.0)*	17.	*Streptomyces* waste	2.78 (6.0)
2.	Cow dung cakes	4.64 (6.0)	18.	*Streptoverticillium* waste	3.52 (6.0)
3.	Groundnut shells	4.62 (6.0)	19.	Wood rotting fungi	4.21 (6.0)
4.	Rice husk	4.68 (6.0)	20.	Bacterial consortium	4.00 (6.0)
5.	Rice straw	4.69 (6.0)	(E)	Algae biomass	
(B)	Industrial waste/by-products		21.	Mixed algae biomass	4.16 (6.0)
6.	Dairy waste sludge	4.71 (6.0)	(F)	Photosynthetic trees/plants waste	
7.	Saw dust	4.64 (6.0)	22.	*Parthenium* sp.	4.22 (6.0)
8.	Sugarcane Bagasse	4.03 (6.0)	23.	*Eichornia* leaves	4.62 (6.0)
9.	Tea powder waste	4.73 (5.0-9.0)	24.	*Eichornia* roots	4.76 (7.0-9.0)
(C)	Municipal solid waste components		25.	*Eichornia* stem	4.66 (6.0)
10.	Nirmalya (Waste flowers)	4.60 (6.0)	26.	Runners	4.67 (6.0)
11.	Compost	3.22 (6.0)	27.	*Tectona grandis* leaves	4.63 (6.0)
12.	Vegetable waste	3.88 (6.0)	28.	*Lantana camara* leaves	2.74 (5.0)
(D)	Fungal and Bacterial waste/biomass		(G)	Reference materials	
13.	*Ganoderma* sp.	4.07 (6.0)	29.	Activated charcoal	5.00 (6.0)
14.	Yeast biomass	3.06 (5.0)	30.	Bagasse Fly ash	4.70 (6.0)
15.	*Mucor heimalis*	2.29 (6.0)		Control (without biomass)	0 (6.0)

Source: Gaddi & Patil (2011); *Values in parentheses indicate optimum pH; All the values in table are average of two readings

Table 3. Silver-cyanide loading capacity of some selected biosorbents

(Bio)sorbent	Without pre-treatment		With pre-treatment
	Conditioned biomass (at optimal pH)	Unconditioned biomass (at original biomass pH)	L-cysteine treated biomass
Eichornia roots	9.74 (7.0)	9.77 (7.01)	13.62 (140%)
Tea powder waste	9.41 (5.0)	9.40 (4.94)	ND
Activated charcoal	9.95 (6.0)	9.94 (5.59)	ND

All the values presented in table are average of two readings; ND: Not Determined

Loading capacity of biomass could be taken as an equivalent measure of binding sites present. Loading capacity values of *Eicchornia* root biomass (9.74 µmol/g), was highly competitive and comparable with activated charcoal (9.95 µmol/g), which was used as reference material (Table 2). This opens up new possibilities of developing an efficient biosorption technology for the recovery of anionic species like and silver-cyanide, gold cyanide and similar other species. These results clearly indicate that many such

screening programmes should be carried out in search of right candidate for efficient sorption of precious contaminants. Table 4 shows the optimum values of DCAG sorption by *Eicchornia* root biomass.

Table 4. Optimum values for maximum biosorption of silver-cyanide (DCAG) by *Eicchornia* root biomass

Sr. No.	Parameter	Optimum value
1.	Temperature	5 - 45°C
2.	Biosorbent quantity	2% (w/v)
3.	Rate of uptake	40 min

Batch studies were also conducted on impact of cations, anions and pretreatment of biomass on biosorption of DCAG by *Eicchornia* root biomass. We also observed that biomass pretreated with L-cysteine enhances the biosorption capacity (Table 3). Biosorption of DCAG by *Eicchornia* root biomass conformed to the Langmuir and Freundlich isotherm models. Continuous mode study in fixed bed column showed that the service time (breakthrough point) obtained for silver-cyanide was 40 hours (data not shown). Thus it could be concluded from our screening/feasibility studies that *Eicchornia* root biomass was an efficient low cost biomaterial for the sorption/removal of silver-cyanide from solutions.

5. An integrated model for the recovery of gold/silver from electronic scrap

In view of above mentioned theoretically sound and practical discussion along with argument and counter argument, the authors here attempt to propose a sound strategic model for the recovery of gold/silver using a combination of hydrometallurgical (chemical) and biometallurgical (biosorption) processes. Fig. 1 shows the broad strategic framework for recovery of precious metals from WEEE. As per this model, the segregated electronic scrap material consisting of gold/silver should first be subjected to hydrometallurgical process using cyanide as a leaching agent. The gold-/silver-cyanide (leachate) thus obtained should then be subjected to biometallurgical (biosorption) process using low-cost biomass (like *Eicchornia* root biomass), wherein the biomass will sorbed the gold-/silver-cyanide within an hour. After sorption and elution using suitable solvent, recovered gold-/silver-cyanide will be obtained. The recovered material then could be used as input material.
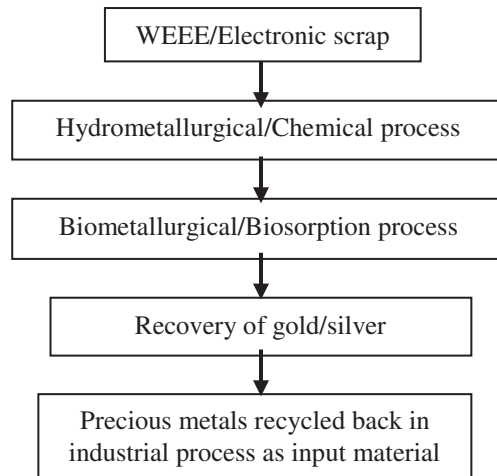

Fig 1. Proposed broad strategic framework for recovery of precious metals from WEEE

Fig. 2 shows the detailed integrated model for recovery of precious metals (gold and silver) from electronic scrap. Prior to leaching of gold/silver from electronic scrap, it is important to thoroughly segregate the different components of WEEE in a well-structured and organised processing station or materials recovery facility (MRF). This involves the steps like separation of functional parts and hazardous materials followed by shredding, screening, magnetic and Eddy current separation of ferrous and non-ferrous materials and density separation.
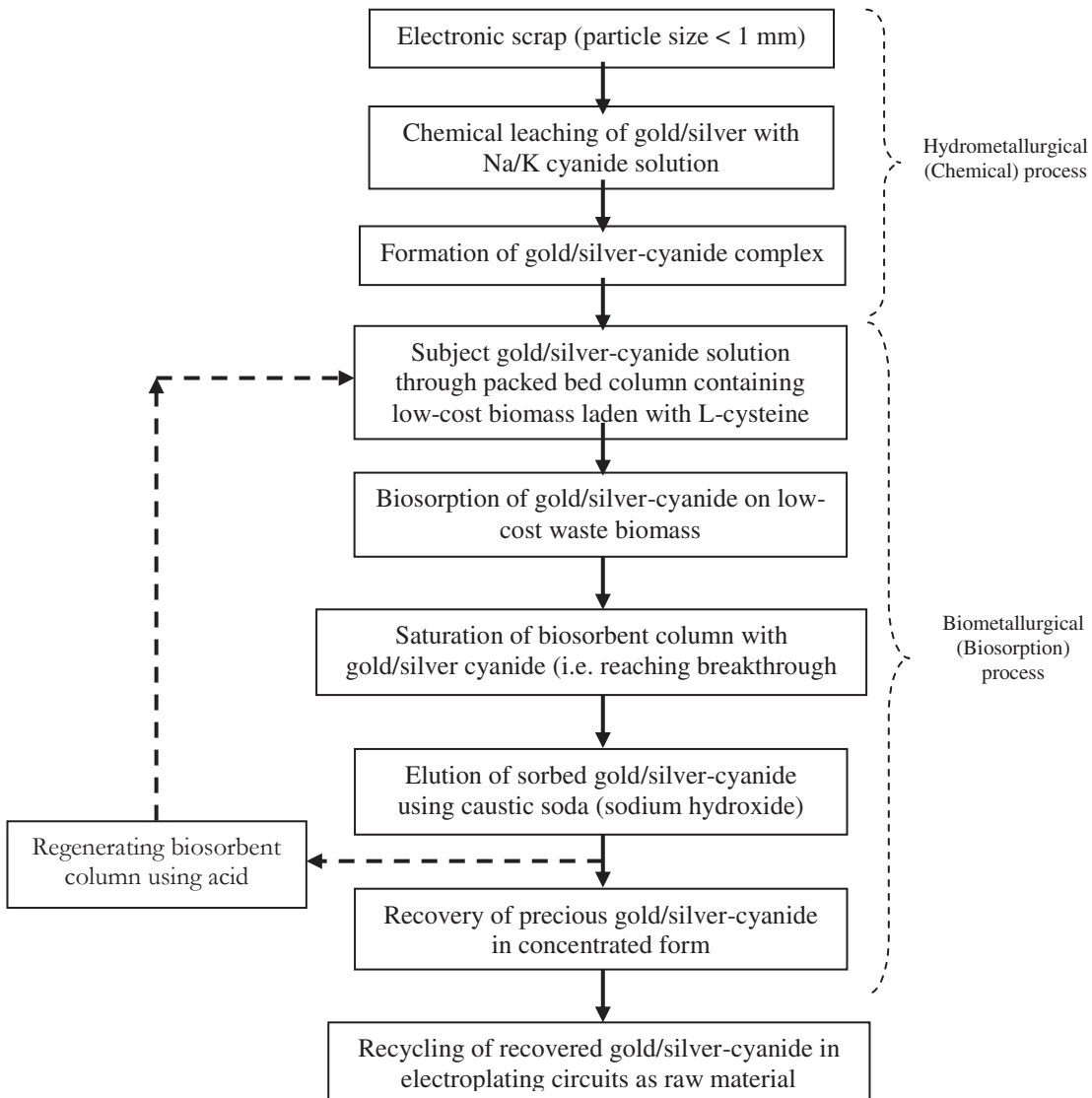
Fig 2: Proposed integrated model for recovery of precious metals (gold and silver) from electronic scrap

In hydrometallurgical process, the electronic scrap of appropriate size (< 1 mm particle size) thus obtained is then leached with cyanide for the extraction of gold and silver. Cyanide treatment ensures the formation of aqueous complex like gold-cyanide and silver-cyanide. Formation of gold- and silver-

cyanide solution after pH adjustment is now ready for its biosorption using selected low-cost biomass. Our biosorption results have shown that the low-cost biomass when treated with a chemical called L-cysteine enhances the sorption capacity of biomass (Table 3).

The biomass ladened with L-cysteine is packed in cylindrical glass column of appropriate length and diameter (also called as packed bed column). The leached gold- or silver-cyanide solution is passed through packed bed column in upflow mode/direction with the required speed. The solution emanated from the column is free of precious metal-cyanides since it has been sorbed on the biomass present in column. The biosorption column gets saturated with gold- or silver-cyanide (after passing several volumes of leached solution) of after specific time period (called its service time or breakthrough point) which completely depends upon the concentration of precious metals and cyanide, size of the column and amount of biomass in it. The biomass loaded with gold- or silver-cyanide is then eluted/leached out by using minimal quantity of suitable eluting agent like caustic soda (i.e. sodium hydroxide). This elution process will help to recover the concentrated form of gold- or silver-cyanide. After complete elution, the biosorbent column could be regenerated using acid in order to use the column for the next cycle of biosorption. The eluted out concentrated form of precious metal-cyanide is now ready for its use as raw/input material for the same industry or any other industry like electroplating industry. Most of the plating industries use silver-cyanide and gold-cyanide baths for plating range of materials.

Generation of e-waste by IT sector, industries, households, commercial and institutional sources on grand scale is completely an urban problem, and is somewhat similar to municipal solid waste (MSW). With the increase in population, industrialisation and other modern practices the usage of electronic goods is bound to increase in future even though the overall size of electronic goods are decreasing. By 2020, the amount of e-waste generation is going to increase five-fold in India making the problem of e-waste management more severe. It is therefore, necessary to formulate innovative strategies/models for effective management of e-waste. Our proposed integrated model is one such effort towards recovery/recycling of precious metal contaminants occurring in WEEE. Furthermore, our model will not only add to the advancement of knowledge in the field of resource conservation and pollution management, but will also strengthen the upcoming discipline of 'Industrial Ecology/Ecosystems'.

Acknowledgements

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