

CHAPTER 1

INTRODUCTION

1.1 Overview

There has been a large amount of attention given to study and research of the alternative lixivants in hydrometallurgical processes for gold recovery from gold scrap. Although a considerable amount of research has been conducted on various leaching of gold, there has not been any commercial introduction of this leaching system.

Leaching is commonly done by using a variety of chemicals which include nitric acid (Krupkova *et al.*, 1987), sulphuric acid, nitric acid and hydrogen peroxide (Kristofova and Karnil, 1996), aqua regia (Lee *et al.*, 1997), ferric chloride (Hanna, 1995; Laatikainen and Paatero, 2005; Vinals *et al.*, 2006), thiourea (Sheng and Etsell, 1998; Becker *et al.*, 1983; Li and Miller, 2002; Whitehead *et al.*, 2009; Parker and Hope, 2008; Li and Miller, 2007; Gonen *et al.*, 2007; Zheng *et al.*, 2006; Li and Miller, 2006), potassium iso-cyanate (Khalemski *et al.*, 1998), potassium iodate and iodine (Leibovitz, 1998), iodide-nitrite mixture (Novoselov and Peshchevitskii, 1998), thiosulphate (Zhao *et al.*, 1999; Feng and Deventer, 2006; Navarro *et al.*, 2002; Chandra and Jeffrey, 2004; Hemmati *et al.*, 1989; Abbruzzese *et al.*, 1995; Senanayake, 2005) and cyanides (Leao and Cimmenelli, 2000; Marsden and House, 1992).

Compared to cyanide, thiourea is less toxic and dissolves gold and silver at faster rates. But thiourea is regarded as carcinogenic and sensitive to the presence of base metals such as copper, lead, zinc, arsenic and antimony (Yavuz and

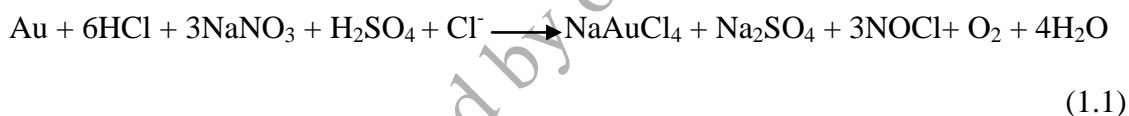
Ziyadanogullari, 2000), which are invariably present in various secondary sources used for gold recovery. The cyanide leaching process commonly adopted in industrialized countries cannot be employed in developing countries due to high risk of exposure to the toxic chemicals.

Thus, there is a need for the development of an appropriate technology for gold recovery from gold scrap, which should have salient features like selectivity, high recovery, economical and eco-friendly even when operated under small-scale conditions (Syed, 2006).

Such scraps and wastes need to be pre-treated before refining to reduce costs and maximise recovery of precious metals. Periodic refining of scrap is important to remove deleterious, embrittling impurities that tend to build up over time and lead to quality problems (Corti, 2002). In any strategy to recover precious metals there is no sense in spending more on processing costs than the value of the metal recovered. The gold purity obtained will vary depending on refining technique and operating skill (Corti, 2006).

1.2 Present invention

This invention relates to the extraction and recovery of gold from gold scraps such as old gold jewellery, gold sludge or gold wastes using a new leaching method differ from prior art in this field since the present invention involves the reaction between sulphuric acid and sodium nitrate (NaNO_3) resulting of nitrous oxide which oxidized gold metal (Au) to form gold ions $[\text{Au}^{3+}]$, then complexed with chloride ions from hydrochloric acid to form sodium tetrachloroaurate (III), NaAuCl_4 . The equation below representing the reaction process:



The present invention provides in one embodiment a reagent for the dissolution of metallic gold comprising solution containing:

- i) Hydrochloric acid which liberates chloride ions to complex with gold ions as tetrachloroaurate (III) .
- ii) Sulphuric acid which reacts with sodium nitrate by generating nitrous oxide to oxidize gold metal to form $[\text{Au}^{3+}]$.
- iii) Sodium nitrate which is selected as an oxidant because it has high potential to effectively oxidized gold when reacts with sulphuric acid.

1.3 Scope and objective of study

The main objective of this study is to find a new hydrometallurgical process that capable to extract gold from gold scrap or alloy. It is our desire to select a gold leaching system that could safely and completely dissolve gold scrap metal and alloy at ambient condition within the certain limit of time without the generation of hazardous by products and with the least production of residuals requiring solidification. To achieve the project objective, these factors are investigated;

- i) Study on the capabilities of ascorbic acid as precipitating agent in the recovery process.
- ii) Investigation yielded on recovery efficiency process and the purity of recovered gold.
- iii) Comparison between this method and aqua regia.

The scope of this study included the effect on the gold dissolution of variables, such as sulphuric and hydrochloric acids concentration, temperature, reaction rate, sodium nitrate, thickness of sample, and selectivity of ascorbic acid as reducing agent.

CHAPTER 2

LITERATURE REVIEW

2.1 Gold properties

Gold is the most noble of the metals being the only one which is not attacked by either oxygen or sulphur at any temperature. Gold is soft, yellow metal with the highest ductility and malleability of any element. Gold has an atomic weight of 196.967 and a density at 20°C of 19.32 g cm³. The pure gold melts at 1063°C and boils at 2966°C. Gold is normally found in nature in its elemental state because it does not bond with most chemical elements. It is opaque and metallic golden-yellow in colour, but it is paler when alloyed with silver in the variety electrum. Most gold is found as tiny grains, platy masses, and nuggets. Gold is widespread in occurrence but it is usually found in minute quantities. It is most abundant in hydrothermal veins and placer deposits (Price & Walsh, 2005).

Gold occurs principally as flakes, scales, or crystals of native metal, in which it is typically alloyed with silver and other metals. More than 90 percent of native gold is alloyed with silver, copper, and iron, in the usual order of importance, and lesser fractions with more than two dozen others metal. Gold forms a complete range of solid solutions with silver, copper, nickel, platinum and palladium. Electrum, which is an important naturally occurring alloy, contain from 18 percent to 36 percent by weight of silver (Smith, 1947).

Gold has been shown to have six oxidation states- +1, +2, +3, +4, +5, and +7. Of these, only the +1 (aurous) and +3 (auric) states are common; the others are rarely observed. Although the complex compound of trivalent gold, ore forming fluids tend to

have low oxidation state in them, and gold is transported mainly in complexes of the type $[\text{Au}(\text{HS})_2]^-$ and $[\text{AuCl}_2]$, along with several other complexes of lesser quantitative importance (Butterman and Amey, 2005).

The beauty and rarity of gold has led to its use in jewellery and in coinage and as a standard for monetary systems throughout the world. Since gold is such a soft metal, it is usually alloyed to give it more strength. In these alloys the term karat is used to express the amount of gold present, 24 karats being pure gold. Alloy of gold metal the other elements of group IB are most frequently used. These coinage gold typically contains 10% copper, while gold used in jewellery and dentistry may contain varying proportions of silver and copper.

The colours of the alloys are frequently controlled by the other metal. Thus an alloy containing 50% copper is dark red, and one containing 65% silver is white and hardly distinguishable to the eye from pure silver. The white gold commonly used in jewellery contain 75-85% gold, with 8-10% nickel and 2-9% zinc, while more expensive white alloys are palladium-gold (90% Au, 10% Pd) and platinum-gold (60% Au, 40% Pt). Gold also has high thermal and electrical conductivity, hence its use in electronics (Renner and Johns, 1989).

Although gold will unite with most metals, comparatively few of alloys are of practical use in the arts or manufactures. The more important alloys of gold are those with copper and silver which are largely used in the jewellery trade and industry. The alloying of gold has in view the hardening of the metal, the lowering of the freezing-point for soldering purposes, and the cheapening of the production; at the same time considerable importance is attached to the colour of the alloys. Advantage is taken of the bleaching action of silver on the yellow colour of gold, to overcome to a great extent the reddening effect of copper. The proportion of gold in these alloys is known as

“fineness” and is expressed either in parts per thousand or in carats (twenty-fourth parts) (Smith, 1947).

2.2 Hydrometallurgy treatment

Hydrometallurgy is the field of extractive metallurgy involving the use of aqueous chemistry for the recovery of the metals from ores, concentrates and recycled or residual materials. Hydrometallurgy is typically divided into three general areas :

- i. Leaching
- ii. Solution concentration and purification
- iii. Metal recovery

In hydrometallurgical treatment, the main steps are acid or caustic leaching of solid material. This process normally requires a small grain size to increase the metal yield. From the solutions the metal of interest are then isolated and concentrated via process such as solvent extraction, precipitation, cementation, ion exchange, filtration and distillation. Leaching solvents are mainly sulphuric acid, hydrogen peroxide, aqua regia, thiourea, cyanide leach solutions, nitric acid, sodium hydroxide, hydrochloric acid etc (Antrekowitsch *et al.*, 2006).

Hydrometallurgical methods also lead to high purity of the metals with the possibility to a selective leaching of the metals in various steps using different solvents. Disadvantages are the high volumes of leach solutions. Furthermore solutions can be corrosive and toxic which lead to high amount of wastes. Metal losses also occur due to composite materials (Antrekowitch *et al.*, 2006).

Hydrometallurgical processes for precious metals recovery from secondary sources generally involve three stages: pretreatment, recovery and refining (Fig.2.1).

Among these, recovery is an important step wherein gold is retained in the elemental state, a process called peeling. Alternatively, dissolution of gold into an ionic state is affected by leaching process.

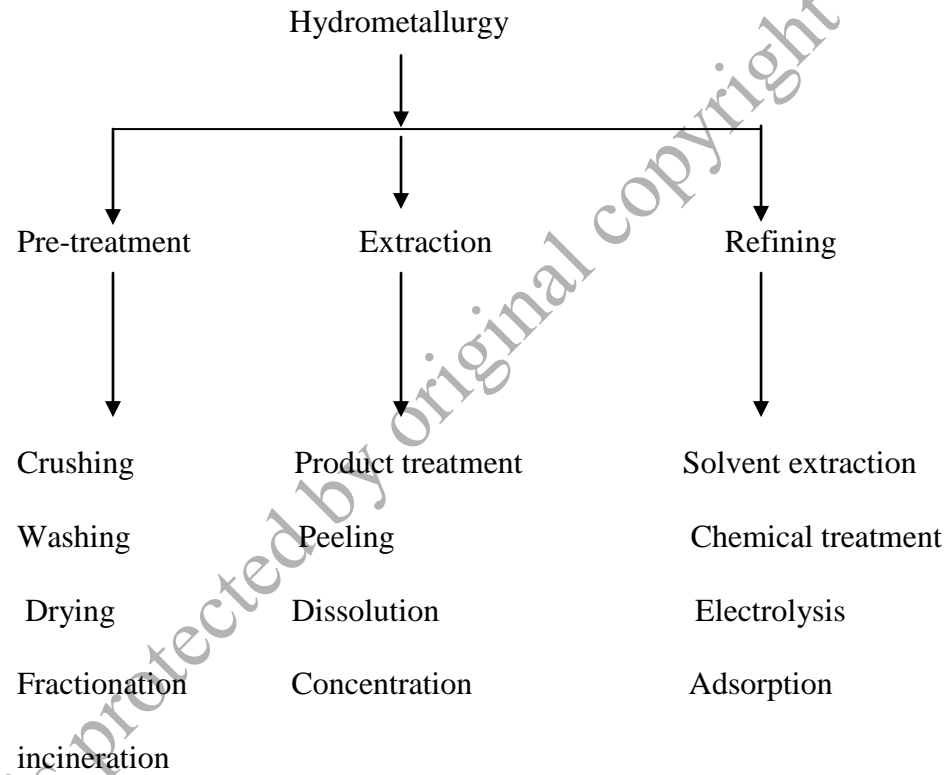


Figure 2.1: Three stages in hydrometallurgy and different processes in each stage (Syed, 2006)

Process innovations in hydrometallurgy have historically taken the form of:

- i) testing new reagents for leaching.
- ii) accelerating the rates of leaching and improving the final extractions.
- iii) offering new options in solvent extraction or ion exchange.
- iv) incremental improvements in electrowinning and refining.
- v) improved methods of making liquid-solid separations that involve better and lower water usage (Peters, 1992).

2.3 Refining technique

Refining is a practice that must be done precisely and methodically to ensure the full recovery of gold as well as an end product that is free of impurities which can lead to quality problems when the metal is reused in production. There are several methods commonly used to recover gold metal such as cupellation, inquartation and parting, Miller chlorination process, Wohlwill electrolytic process, Fizzer cell, solvent extraction, aqua regia process and pyrometallurgical process. Some operations are suitable for use by manufacturers and jewellers who wish to refine in-house while others are designed for commercial refiners who handle large lots (Corti, 2002).

The aqua regia process can produce gold of up to 99.99% purity. It is based on the fact that aqua regia (a mixture of hydrochloric and nitric acids in 4:1 ratio) can dissolve gold into soluble gold chloride. The process is most suited to medium to large scale operations. The main limitation of the process is that the feed material should have a silver content of 10% or less to avoid blocking up dissolution of the scrap. Because of this, pre-treatment by the inquartation process to reduce the silver content may be necessary. Alternatively, the low silver content may be achieved by a judicious blending of batches of scrap. Thus, the process is more suitable for medium and high karat gold scrap refining. (Corti, 2006).

2.4 Hydrometallurgy of gold

The hydrometallurgical leaching of native gold from gold-bearing ores or the dissolution of gold metal during the recycling of electronic and precious metal scrap is performed every day using hazardous chemicals such as sodium cyanide or aqua regia.

These chemicals represent health and safety risks for workers and a serious threat for the environment. However, even if several other reagents are known to dissolve gold at the laboratory scale such as thiosulphate solution and thiourea none of these are used industrially (Geoffroy and Cardarelli, 2005).

Gold will dissolve in aqueous solutions containing a good ligand for gold and an oxidizing agent, but neither condition alone is sufficient. Thus, gold will not dissolve appreciably in hydrochloric or nitric acid but dissolve readily in aqua regia, a mixture of three parts concentrated hydrochloric to one part concentrated nitric acid to give tetrachloroauric(III) acid, HAuCl_4 (Svehla, 1982). The reaction involved is as follows:



Similarly gold will dissolve in hydrochloric acid in the presence of hypochlorite or ferric as oxidant. The dissolution of gold in cyanide solutions with air or hydrogen peroxide as oxidant is another example.

The cyanidation process has been used to leach gold over 100 years since it was patented in 1888 by Mac Arthur and Forest brothers (Senanayake, 2004). The reaction with oxygen as oxidizing agent apparently takes place by adsorption of oxygen onto gold surface, followed by reaction of this surface layer with cyanide to first give AuCN and then the complex $[\text{Au}(\text{CN})_2]$ which passes into solution. The dissolution of the metal is given by chemical reaction as follows :



La Brooy *et al.*, (1994) reported that despite the long term use of cyanidation there is a growing interest in non-cyanide gold technology based on the lixivants mainly due to the failure of cyanidation to extract gold from the so-called difficult to treat ores and the environmental and safety issues.

In recent years, as the environmental problems become more and more serious, a lot of countries such as USA, Japan, Turkey and the Czech Republic (De Voto and McNutty, 2000; Damsell, 1998; Midwest Treaty Network, 2001), have banned the use of cyanide, the most commonly used lixiviant for gold. A great deal of investigations (Alymore and Muir, 2001; Feng and Van Deventer, 2002; Fleming *et al.*; 2002; Bruer and Jeffrey, 2000; Jeffrey *et al.*, 2001; Jeffrey, 2001; Ji *et al.*, 2001; Hiskey and Atluri, 1988; Zipperian *et al.*, 1988; Berezowsky and Sefton, 1979; Yen *et al.*, 1996; Fujita *et al.*, 1997; Umetsu and Tozawa, 1972; Wan *et al.*, 1994; Wan, 2002) have been carried out on gold leaching with ammonium thiosulfate as substitute for cyanide (Kejun *et al.*, 2004).

Thiosulfate leaching can be considered a non-toxic process, and the gold dissolution rate is faster than conventional cyanidation. Due to the decreased interference of foreign cations, high gold recovery can be obtained from the thiosulfate leaching. In addition, thiosulfate is cheaper than cyanide. For leaching to occur at reasonable rate, thiosulfate, ammonia, and copper must be present. The chemistry of the ammonia-thiosulfate system is complicated by the presence of complexing ligands such as ammonia and thiosulfate. Thiosulfate stabilizes gold in solution while copper and ammonia accelerate the leaching reaction (Abbruzzese *et al.*, 1995).

In addition, thiosulfate is prone to oxidative degradation. Thiosulfate is meta stable and will eventually decompose in aqueous solutions. However, by maintaining suitable concentrations of the thiosulfate, ammonia, copper and oxygen in the leach

solution, and consequently, suitable activity of electrons (Eh) and pH conditions, thiosulfate leaching can be practical (Molleman and Dreisinger, 2002).

Generally, the thiosulfate leaching conditions reported in the literature (Zipperian and Raghavan, 1988; Navarro *et al.*, 2002) are critical with high reagent consumption. Further investigations are required on leaching under low reagent concentrations over an extended period where reagent consumption is low.

Although there are many reports on the reaction chemistry of gold and thiosulfate, the variables affecting gold dissolution efficiency, thermodynamics and kinetics of gold dissolution with ammonium thiosulfate, gold recovery by cementation and gold recovery by ion exchange resins, there is little information on the extraction of gold from ammonium thiosulfate pregnant solution by solvent extraction (Kejun *et al.*, 2004).

Hot mixtures of hydrochloric acid with strong oxidizing compounds are known to generate in-situ nascent chloride which is capable of dissolving gold efficiently. One of examples is a hot mixture of hydrochloric acid and ground manganese(IV) oxide to dissolve gold metal either under atmospheric or pressurized conditions. The best result was obtained under a pressure of 639 kPa at 90°C with a dissolution rate of 0.250 g.cm⁻²h⁻¹ (Geoffroy and Cardarelli, 2005). The chemical reaction involved during the dissolution of gold in this media is shown below:



Thiourea has been discussed (Deschenes and Ghali, 1988; Chen *et al.*, 1980) as a reagent for precious metals leaching by many authors due to its high complexing ability. Soviet reseachers have reported that thiourea is very effective in the recovery of

noble metals. This has been done in hydrochloric acid, sulfuric acid, and nitric acid media. Thiourea leaching of a gold concentrate is possible without the addition of any oxidant but gold extraction is only 80% in 8 hours leach. The use of hydrogen peroxide is better than ferric chloride, leading to lower thiourea consumption and less impurity build up.

The dissolution of base metals from gold-containing materials in acidic thiourea solution causes not only obstacles against selective leaching of gold but also unnecessary consumption of thiourea due its simultaneous complexation with base metals. Orgul and Atalay (2002) reported that an alkaline solution has been suggested for use in gold leaching media, since many common base metals like copper, nickel, and iron are chemically passivated. However, high reagent consumption, low efficiencies and the poor stability of thiourea are the main obstacles to common use of thiourea for leaching of precious metals.

2.5 Gold Recovery

Gold recovery from aqueous solutions has received significant attention because gold is present in appreciable amounts in electronic parts and plating materials (Nakajima *et al.*, 2003; Ishikawa *et al.*, 2002; Ogata and Nakano, 2005). The increase in the industrial demand for gold will correspond to the increase in the need for gold recycling. In general, gold is separated and purified from industrial wastes by hydrometallurgical processes involving chloride media (Iglesias *et al.*, 1999).

Gold has mostly been extracted from its ores by cyanidation in the Carbon-In-Pulp process. The gold bullion produced from this process generally has to be further refined in mint shops to achieve 99.99% purity (Marsden and House, 1992).

Conversely, gold from scraps is mostly recovered via gold chloride complexes produced from aqua regia digestion. High purity gold requires more elaborated processing, using 99.99% gold as starting material, which is first dissolved in high purity acid and then re-plated. The dissolution of gold in chloride systems has been well-studied, with the gold chloride complex being very stable in solutions of strong chloride, acid and high solution potential (Tran and Davis, 1992; Tran *et al.*, 2001).

Many sorbents and extractants have been proposed for gold recovery, including activated carbon (Navarro *et al.*, 2006; Jia *et al.*, 1998), persimmon tannin gel (Nakajima *et al.*, 2003; Nakajima and Sakaguchi, 1993), ion exchange resins (Warshawsky *et al.*, 2000; Kautzmann *et al.*, 2002; Dally *et al.*, 1996; Zhang and Dreisinger, 2004; Habashi, 1969), oxygenated extractants, thio extractants, ammonium extractants and phosphorus extractants (Das and Bhattacharyya, 1976; Liu *et al.*, 2004). Cox, (1992); Edward and te Riele, (1983) reported that dibutylcarbitol (DBC) and methyl isobutyl ketone (MIBK) are commercially used as extractants for Au(III) in HCl solutions at Inco and Matthey Rustenberg Refiners, respectively. However, they seem to be insufficient in term of their hydrophobicity and selectivity of Au(III). Other less effective extractants used for gold extraction include tributyl phosphate (TBP), 2-ethyl hexyl phosphonic acid mono-2-ethylhexyl ester (PC-88a from China) and tributyl phosphine oxide (TBPO), as reviewed by Shen and Xue (2007).

Monoamide compounds, such as N, N-di-n-octylacetamide (DOAA) and N, N-di-n-octylauramide (DOLA), have also been investigated for the extraction of gold and other precious metals from chloride solutions (Narita *et al.*, 2006). Both DOAA and DOLA provide good extraction systems, which are applicable over a wide range of HCl concentrations (0.1- 4 M), with high selectivity for gold, although the gold loading is higher with respect to DOAA (at 0.5 M), where solution of 90 g/L Au could be obtained

compared to 80 g/L Au for 0.5 M DOLA. The stripping of gold from the organic to the aqueous phase for recovery however, is not complete. Furthermore, the study of gold extraction from chloride media with monoamide compounds is limited (Baroncelli *et al.*, 1988; Gasparini and Grossi, 1986; Pohlandt and Fritz, 1979).

Tasdelen *et al.*, (2009) have investigated gold recovery using diethylaminoethyl cellulose (DEAE-cellulose), a common biopolymer derivative, from synthetically prepared diluted gold-bearing solutions of 50 ppm. It was demonstrated that gold recovery efficiency increased with an increasing amount of sorbents, as well as increasing contact time. A gold recovery efficiency of 99% was attained under conditions of 20-40 g DEAE-cellulose per liter at a shaking rate of 130 rpm for 30 min at room temperature. On the other hand, with smaller amounts of sorbent (6 g L^{-1}), it was also possible to recover gold from the solution with 99% efficiency when the reaction temperature was increased to 60°C .

Using a liquid membrane, Fu *et al.*, (1995) evaluated a technique for recovering gold from a gold-palladium-platinum acidic chloride solution. The extraction of gold chloride from this solution was completed by contacting the leach liquor with trioctylamine-kerosene solution, modified with 20% v/v octanol, at ambient temperature. The separation of gold from the other metals was improved with increasing HCl concentration (0.1- 4.0 M) in the feed solution. The gold from the organic phase was then stripped using HClO_4 . Using a 1 Mm trioctylamine and 20% octanol in kerosene as the extractant, more than 95% of the gold was recovered, leaving 99% of both platinum and palladium in HCl feed after 7 hour.

An improved process was developed to produce to produce gold with purity $>99.9998\%$ from gold scraps using dibutyl carbitol (DBC) as an extractant (Jung *et al.*, 2009). Gold extraction based on DBC is both cost effective and an environmentally

friendly metal recovery process. DBC has a high flash point, low stability in water and low volatility (boiling point 254.6°C). This chemical family has been found to be superior to other basic solvents, such as tri-n-butyl phosphate (TBP) and tri-n-octyl phosphine oxide (TOPO), due to its high selectivity for gold. Distribution ratios (ratio of organic phase gold concentration/aqueous phase gold concentration) between 5000 and 10,000 could be easily achieved during extraction using concentrated HCl (>5 M) in the aqueous feed (Morris and Khan, 1968).

As a result, several patents on process steps based on DBC have been lodged (Lea *et al.*, 1983; Yoshifumi and Takehiko, 1999). Typically, more than 99% of the gold from leach liquors (0.50-1.75 M HCl) could be extracted into DBC, achieving concentrated solution of up to 50 g/L Au, while other metal contaminants exist at mg/L levels. The DBC was then scrubbed several times with HCl (0.25-0.75 M), followed by centrifugal separation, yielding high purity from which 99.998% Au powder could then be recovered by reductive-precipitation using oxalic acid. Lea *at al.*, at Inco (1983) have also patented a solvent extraction process to recover gold using DBC, and palladium /platinum using alkyl sulphide and TBP. For gold, a 99.99% product could be recovered using oxalic acid as reductant. The developed process is shown in Fig. 2.2.

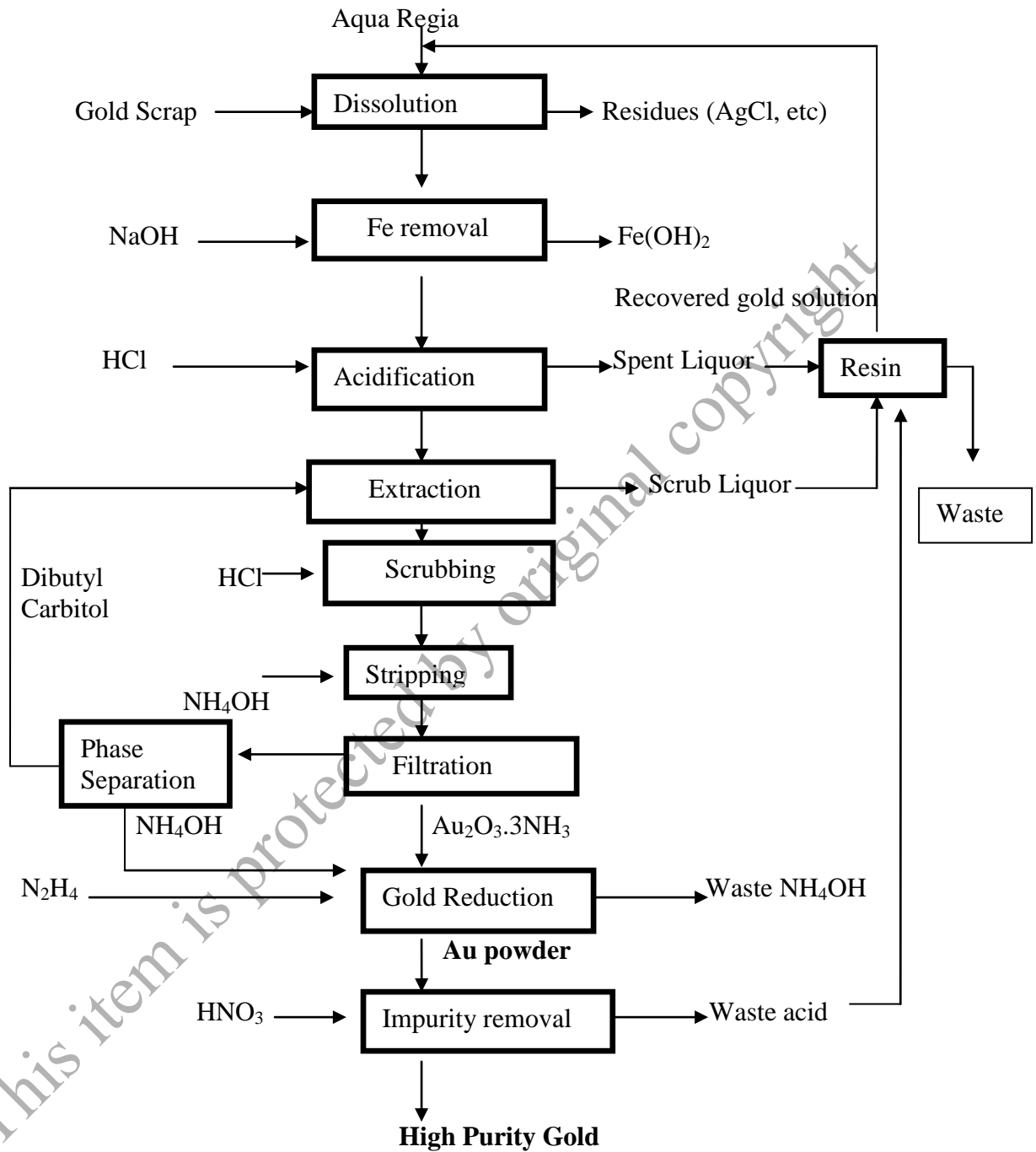


Figure 2.2: Process for producing high purity from scrap materials using DBC (Jung *et al.*, 2009).

2.6 Sources of gold scraps in jewellery manufacture

Scrap arises at all stages in the manufacture of jewellery at the alloying and ingot stage, during stamping and blanking operations, in investment casting (feeders, defective castings, etc.), filing, drilling, sawing and soldering at the workbench, and finishing (diamond cutting, polishing, etc). Scrap comprises redundant material discarded in manufacture and defective items of jewellery that are rejected. This can be high to medium grade in term of precious metal content . The amount of scrap generated can be controlled through careful process design, material utilisation and implementation of best practice on the shop floor, but it cannot be eliminated (Corti, 2002).

Wastes can also arise at each stage of manufacture and and typically low grade in terms of precious metal content. Roeland Loewen (Corti, 2002) has reported typical gold contents of scraps and wastes, as shown in Table 2.2. Clearly, the gold and other precious metal content will depend on the caratages of jewellery being made and type of equipment in use. Platinum group metals may also be present if white golds, dental alloys and platinum jewellery are made on the same equipment. These have value, too, and may be worth recovering separately.

Table 2.2 : Typical gold content for scraps and wastes (Corti, 2002)

Material	Gold content, wt %
Old Jewellery	39-73
Bench scrap	19-52
Sink trap setting	6-8
Carpets and wood floors	0.1-9
Old crucibles	0.8-5
Polishing dusts and sweeps	0.5-5
Watchbands and miscellaneous gold-filled scrap	0.25-5
Emery papers, floor and other shop dirt, brushes	0.1-4

2.6.1 Treatment of scraps and wastes

In any strategy to recover precious metals such as gold, it is important to consider the cost of recovery. There is no sense in spending more in processing costs than the value of metal recovered. In particular, the processing of wastes to reduce bulk and concentrate the precious metal is essential to economic recovery. Refining costs are usually based on gross weight shipped in, if for example, waste is sent to a toll refiner.

Whether directly recycling high grade scrap or sending materials for refining, there are some broad guidelines that should be followed if efficient, cost effective is to be achieved :

- Segregate scrap by colour and by caratage

- Use only clean, high-grade scrap of known composition. Ensure contaminations such as entrained investment, abrasives and inclusion, surface oxides and grease are removed by suitable cleaning procedures such as sand or water-blasting, acid pickling or alkali treatment, and ultrasonic degreasing.
- It is preferable to remelt scrap under a flux cover and grain it before re-use. The use of grain assists melting and alloy homogeneity.
- Always check gold content and alloy composition before adding scrap to new metal to ensure the new melt will meet fineness specification and does not contain deleterious impurities. Scrap from external sources may contain embrittling levels of metals such as silicon and lead or even cadmium-containing solders.
- Do not use more than 50% scrap in a new melt and preferably less, especially in investment casting. The yield of good castings reduces as scrap content of the new melt increases. However, it is important to keep scrap used in balance with scrap generated so it does not accumulate (Corti, 2002).

CHAPTER 3

EXPERIMENTAL TECHNIQUES AND MATERIALS

3.1 Materials and chemicals

Gold strip (99.9% Au, thickness 0.2 mm) with a surface area of about 2cm² was used. These was washed with acetone twice and rinsed with distilled water and dried with lint-free paper. Analytical grade of hydrochloric acid (37%) and sulphuric acid (95-97%) were obtained from Fischer, sodium nitrate (99.9%) and ascorbic acid (99.7%) were obtained from Q Rec and Scharlau, respectively. Distilled water was used in the experiments.

3.2 Instrumentation

Following each recovery process, the concentration of gold ions was measured with atomic absorption spectrometer (AAS) model GBC 322 Plus. Nitrates presence in the filtrate were determined with ultraviolet- visible spectrophotometer (UV) model Carey. The particle size and composition of the gold precipitate were analyzed with scanning electron microscopy (SEM) EDAX model Carl Zeiss Supra 40 VP and qualitative analysis for gold scrap was determined with X-Ray Fluorescence Spectrometer model Bruker S4 Pioneer.

3.3 Experimental procedure for making gold strip

Gold scraps (containing approximately 85-90% Au) from various sources were collected and dissolved into aqua regia (HCl : HNO₃ of 3 : 1 volume ratio) at 90-100° C, using Pyrex glass beaker equipped with heating control. The solution was filtered and washed 5 times with 20 mL portions of distilled water and the residue set aside. The filtrate was evaporated on a hot plate until it had a syrupy consistency. A few drops of concentrated hydrochloric acid were added to it, and the solution diluted with 50 mL of distilled water before being evaporated again to a syrupy state. This process of evaporation, dilution and re-evaporation with the addition of HCl was found to be crucial step in the recovery process and was repeated at least 3 times. The excess nitric acid was completely removed when there was no brown fumes appeared in the solution (Smith, 1947).

The solution was acidified with 5 mL of concentrated hydrochloric acid and a few drops of sulphuric acid before being diluted to 150 mL of distilled water. The solution was covered with a watch glass and 25 mL of saturated oxalic acid solution was added and the mixture was boiled on the hot plate. The gold that precipitated was filtered off, washed with hot water and dried at 110°C for 1 hour. The dried brown gold powder was mixed with an equal mass of lithium tetraborate flux and placed in a clay crucible. This was melted at 1063°C in the muffle furnace (Fig. 3.1). After melting, the flux was separated from the gold by using hammer and screw driver. Then it was rolled to form gold strip using a rolling machine to a thickness approximately 0.20 mm (Fig. 3.2).



Figure 3.1: Gold melted at 1063°C in a clay crucible.



Figure 3.2: Gold strip after rolled with rolling machine.

3.3.1 Leaching tests

Leaching tests were performed in Pyrex glass beakers, covered with watch glass on the hot plate with temperature indicator display. Approximately 1.0 g of gold strip which cut into small pieces (1.0 cm^2) was weighed and put into 400 mL beaker which initially charged with 40 mL of concentrated hydrochloric acid. 3.0 g of sodium nitrate (0.64 M) was added into a beaker followed by 15 mL of sulphuric acid. The reaction was observed for 2 hours. The sample was then removed from the reagent, washed, filtered with Whatman® filter paper No. 40, dried and re-weighed to determine the weight loss. This test was repeated with various concentration of sulphuric acid (1 to 18 M) and hydrochloric acid (1 to 11.5 M) in each leaching tests and temperature was varied from 50°C to 200°C .

3.3.2 Calculation of gold dissolution percentages

The initial (before leaching test) and final weights were denoted w_1 and w_2 respectively. The weight loss resulting from the dissolution of a sample in the acid solution was calculated using the following equation,

$$\Delta w = w_1 - w_2 \quad (3.1)$$

The gold dissolution after 2 hours then can be expressed as follows,

$$\% \text{ Au} = \frac{\Delta w}{w_1} \times 100\% \quad (3.2)$$