



Recovery of precious metals from ammoniacal thiosulfate solutions by hybrid mesoporous silica: 3 – Effect of contaminants



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ABSTRACT

Precious metal concentration and/or separation processes based on adsorption such as in hydrometallurgical systems are often affected by the presence of dissolved contaminant/impurity species. This research is the first report on investigation of the effect of impurity metal and oxyanion associations with gold and PGM concentration by hybrid mesoporous silica adsorbent in neutral-pH simulated thiosulfate leach solution. It was found that the increased solubility of Fe(III) or Cr(III) salts beyond a certain loading limit was accompanied by a decrease in the equilibrium pH. The latter adversely affected precious metal recovery by impeding adsorption (of platinum and gold) and causing precipitation (of palladium) in the thiosulfate solutions. Increasing loading concentration of softer Lewis acid cations such as those of nickel, zinc and lead caused gradual decrease of the precious metal adsorptions with minor effects on the pH. While platinum adsorption recovery was least affected by up to 200 mg/L nickel loading, gold adsorption was almost unaffected by up to the same amount of lead addition. Arsenic oxyanions were almost fully dissolved while antimony and tellurium salts were mainly insoluble in the thiosulfate equilibrium system. Increasing total oxyanion loading concentration (up to about 1500 mg/L) resulted in gradual rather slow decrease in gold and PGM adsorption recoveries. In the end, highly selective adsorption of precious metals by both thiol and amine organic functional groups on hybrid mesoporous silica was achievable provided that concentration of metal/oxyanion contaminants were controlled and pH was further stabilized at the 7–8 regime.

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1. Introduction

Thiosulfate leaching has been well recognized through the developments toward cleaner and safer gold ore processing using hydrometallurgy. This is while more traditional techniques using reagents such as cyanide and halides have been proved to be more aggressive, unsafe both in operation and environment and costly with respect to both protection and disposal issues. Thiosulfate anion is known to form relatively strong complexes with precious metals such as gold and silver over a relatively wide range of pH and E_h values. It has also been established that the presence of ammonia in the system hinders the dissolution of iron oxides, silica, silicates and carbonates, the most common *gangue minerals* found in gold-bearing ores [1,2]. Other than metal mining, thiosulfate leaching has been considered for recovering precious metals from secondary sources such as electronic devices as part of the overall recycling procedure [3,4].

Natural deposits of primary or secondary value precious metals always contain minor to significant contents of other metallic minerals and compounds. The most frequent and/or abundant metallic associates in gold/silver and PGM (platinum group metals) resources are summarized in Table 1. Among common impurities the so-called “iron-group elements” (arguably ranging from Cr through Ni), are repeatedly reported to be associated with mining and processing as well as recycling operations of gold and especially PGM. There could be secondary sources of contamination such as the separation and process equipments that might introduce iron species (including metallic iron) into the concentration and separation system. It is known [14] that in gold leaching when pH values are lower than 8.0, metallic iron (from grinding media) and iron salts begin to dissolve considerably in solution, adversely affecting gold dissolution from minerals. Electrochemical reduction of iron from ferric, Fe(III), to ferrous, Fe(II), state is associated with oxidation of thiosulfate species in solution.

Chromium occurrences in natural gold deposits, on the other hand, are not as common. Nevertheless, the interests in the current research to investigate the effects of dissolved chromium on the adsorption system arises from the chromium associated with

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PGM extraction. Problems were reported in some enrichment processes of PGM-bearing chromite ores such as in the “UG-2” section of the Bushveld complex in South Africa. It has been indicated that the presence of trivalent Fe and Cr oxides (spinel mineralogy) in the secondary cleaner flotation concentrate have caused low recovery of precious metals and higher operating costs using the conventional smelting of the PGM-bearing concentrates. Combinations of roasting followed by HCl/chlorine leaching and ion exchange separation [15] as well as cyanide and bio-leaching [16] were proposed to improve recovery of PGM metals in such concentrates.

Besides occurring as accessory or impurity-level contaminants in precious metal deposits, lead and zinc when are mined and processed from major base metal deposits, could end up producing low grade sulfide flotation tailings. Such primary tailings material could become the target of certain recycling/scavenging processes for the precious metal values that could also turn out high precious metal recoveries in some mineral processing operations [17–19].

Despite high occurrence of the semimetal elements such as As, Sb and Te in many of the natural ores including precious metal deposits, generally, information on their possible interference with leaching, purification and further processing and disposal stages are rare. One possible reason has been weak (or no) interaction/complexation of dissolved As and Sb species with cyanide, $[\text{CN}]^-$, to be regarded problematic in the more recognized and practiced gold cyanidation industry. Usually, the solubility of these metals from their parent minerals to form oxyanions in the leaching solutions has a strong dependence on pH, and normally in non-acidic pH below 10, they are known to remain mainly as insoluble solids [20]. However, there are several parameters (e.g. oxidation by ferric ions) in more complex solution chemistries that could render such metals to dissolve and possibly interfere with both the leaching/concentration processes. For example, it is known [21] that the lower oxidation forms of the oxyanions (such as arsenite and antimonite) are oxygen consumers so they might modify the oxidation potential and therefore speciation in the system.

There have been several researches devoted only to study the effect of such impurity species during leaching stage of the gold hydrometallurgy including the effect of iron species [22,23], heavy metal ions [24], multiple metal ions in complex sulfide ores [25] as well as the adverse effect of metallic minerals (sulfides and oxides) [26]. Despite several different and sometimes repetitive separation techniques prior to leaching, many of the unwelcome species would finally find their way into the leach solution and therefore would interfere with the solution purification and concentration stages as well.

The concentration step which follows the leaching in the hydrometallurgical processes usually involves the separation of precious metal compounds from the rest of the leach solution. There has been substantial amount of research on the use of anion exchange resins [17,27–29] to recover and concentrate gold and silver from thiosulfate leach solutions, whereas the adsorption on activated

carbon and cementation methods have not generally found to be technologically/economically efficient for gold thiosulfate system. Moreover, since other contaminant metals such as lead and copper could also be leached/dissolved to form complexes with thiosulfate, competitive adsorption would be a significant issue if not an impediment for similarly charged anionic species to ion exchange on the resin surface sites. In such cases, limiting the contact time of the adsorbent with solution and use of more dilute solutions have been recommended [29,30]. On the other hand, non-carbon adsorption systems such as mesoporous silica adsorbents have still been rather novel to the industry. The published research on the precious metal recovery from single or multiple component solutions have mainly been concerned with the acidic systems [31–37].

The current study is a continuation of research by the same authors on gold and PGM adsorption in thiosulfate system by hybrid mesoporous silica. This work is specifically devoted to study the separation of precious metals by adsorption in presence of different concentrations of the mentioned impurities in a simulated thiosulfate leaching system. In this regard, functionalized MSU-2 mesoporous silica material was used as adsorbent. The MSU-X family of mesoporous silicas, in general, and MSU-2 in specific, are recognized with their characteristic wormhole inter-connected random pore channels in a 3D distribution that could provide high accessibility of adsorbate species to the functional (ligand) sites on the mesopore walls [39,40]. Simple functional organic ligands (of thiol, $-\text{SH}$, and amine, $-\text{NH}_2$ type) were used to functionalize the surfaces of the mesopore walls in the silica framework. These functional sites will be responsible for (predominantly *specific*) adsorption of precious metal species from solutions.

2. Materials and methods

2.1. Chemicals

A list of all different chemicals used both in preparation of adsorbent material and in adsorption tests are provided in Table 2. All other reagents such as acids, bases and solvents and oxidizing agent (H_2O_2) were reagent grade.

2.2. Hybrid mesoporous silica adsorbent and its properties

Mesoporous silica (of MSU-2 type) was prepared through sol-gel synthesis benefiting from the neutral self-assembly route with little modifications of the methods adopted by researchers at Michigan State University (USA) as well as at Laboratoire des Matériaux et Procédés Membranaires (France) [40–43]. The mesopore walls of the silica framework were functionalized using either or both of thiol and amine functional organosilanes during and after condensation of the silica framework respectively. The as-synthesized and functionalized mesoporous structures were examined

Table 1
The most common associated metals in the processing and recycling of precious metals.

Precious metal source	Origins of the impurity ions/compounds	Major metallic elements that could end up being dissolved in the aqueous medium	References
Primary natural (hard-rock) ore deposits	Tellurides, sulfides, antimonides and arsenides of precious metal(s) and other metals as well as their oxidation products	Au, Ag Cu, Fe, As, Sb, Te, Hg, Bi, Pb, Zn, Cd, W, Mo PGM (Pt, Pd, Rh, Ru, Ir, Os) Cu, Ni, Fe, Ti, V, Cr, Bi, As, Sb, Te, Se, Co, Zn, some REEs	[5–9]
Recycling streams	Major sources among others: <ul style="list-style-type: none"> • Process catalysts and autocatalysts • Electronics mainly printed circuit boards (PCBs) • Jewellery • Medical 	Vary depending on the source, generally include but not limited to: Au, Ag, PGM, Si, Al, Mg, Fe, Cu, Ni, Sn, Zn, Mn, Cr, Ce, Zr	[10–13]

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