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Process simulation and gate-to-gate life cycle assessment of hydrometallurgical refractory gold concentrate processing

Heini Elomaa¹ · Pia Sinisalo² · Lotta Rintala³ · Jari Aromaa¹ · Mari Lundström¹

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Abstract

Purpose Currently, almost all cyanide-free gold leaching processes are still in the development stage. Proactively investigating their environmental impacts prior to commercialization is of utmost importance. In this study, a detailed refractory gold concentrate process simulation with mass and energy balance was built for state-of-the-art technology with (i) pressure oxidation followed by cyanidation and, compared to alternative cyanide-free technology, with (ii) pressure oxidation followed by halogen leaching. Subsequently, the simulated mass balance was used as life cycle inventory data in order to evaluate the environmental impacts of the predominant cyanidation process and a cyanide-free alternative.

Methods The environmental indicators for each scenario are based on the mass balance produced with HSC Sim steady-state simulation. The simulated mass balances were evaluated to identify the challenges in used technologies. The HSC Sim software is compatible with the GaBi LCA software, where LCI data from HSC-Sim is directly exported to. The simulation produces a consistent life cycle inventory (LCI). In GaBi LCA software, the environmental indicators of global warming potential (GWP), acidification potential (AP), terrestrial eutrophication potential (EP), and water depletion (Water) are estimated.

Results and discussion The life cycle assessment revealed that the GWP for cyanidation was 10.1 t CO₂-e/kg Au, whereas the halogen process indicated a slightly higher GWP of 12.6 t CO₂-e/kg Au. The difference is partially explained by the fact that the footprint is calculated against produced units of Au; total recovery by the halogen leaching route for gold was only 87.3%, whereas the cyanidation route could extract as much as 98.5% of gold. The addition of a second gold recovery unit to extract gold also from the washing water in the halogen process increased gold recovery up to 98.5%, decreasing the GWP of the halogen process to 11.5 t CO₂-e/kg Au. However, both evaluated halogen processing scenarios indicated a slightly higher global warming potential when compared to the dominating cyanidation technology.

Conclusions The estimated environmental impacts predict that the development-stage cyanide-free process still has some challenges compared to cyanidation; as in the investigated scenarios, the environmental impacts were generally higher for halogen leaching. Further process improvements, for example in the form of decreased moisture in the feed for halide leaching, and the adaptation of in situ gold recovery practices in chloride leaching may give the cyanide-free processing options a competitive edge.

Keywords Cyanide-free · Gold leaching · Gate to gate · Halides · Pressure oxidation · Simulation

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1 Introduction

Cyanidation of gold has dominated the gold leaching industry since the 1900s as a generally effective and fairly low-cost process. However, direct cyanidation of refractory ores, low-grade ores, and certain complex ore types suffer from low gold recoveries (< 80%), high cyanide consumption, or both. In addition, the toxicity of cyanide and related strict environmental legislation control the use of cyanide and operations in the gold plant (Fleming 1992). These factors motivate not only the development of cyanide-free gold processes but also their early-stage evaluation of process mass and energy balances as well as environmental impacts.

In the recent decades, 27 different lixiviants have been investigated as possible replacements to cyanidation, most of them based on thiosulfate, thiourea, and halide systems (Aylmore 2016). More recently, cyanide-free processes using chloride solutions at ambient pressure and at low temperature have been researched and tested on pilot scale (Ishiguro et al. 2008; Haavanlammi et al. 2010; Harris and White 2014). In halogen/halide processes, the oxidant can be chlorine (Rose 1898), hypohalides (Yen et al. 1990), a cupric/cuprous redox couple (Lundström et al. 2014; Lundström et al. 2015; Lampinen et al. 2017), or a ferric/ferrous redox couple (Seisko et al. 2018) and the complexing agent is chloride. Many literature reviews have explored the chemistry of these alternative processes (Nicol et al. 1987; Tran et al. 2001; Hilson and Monhemius 2006; Aylmore 2016), and it is generally acknowledged that the kinetics in halide solutions are faster compared to cyanidation (Jeffrey et al. 2001). Gold leaching by hypochlorite has been studied by Baghalha (2007) and Yen et al. (1990), who suggest that an oxidative pretreatment should be adopted when processing refractory ore (Yen et al. 1990). Gold leaching in hypohalide media is highly dependent on the acidity and generates chlorine gas (Lalancette 2009; Lalancette et al. 2015a). A halogen process utilizing this phenomena has been researched by Lemieux et al. (2014) and De Michelis et al. (2013). Both these studies show that the hypohalide system can provide efficient gold leaching both on laboratory and demonstration scale.

An increasing amount of global gold production derives from refractory-type materials. These materials have high concentrations of sulfide minerals that encapsulate the gold, such as pyrite and arsenopyrite, and preg-robbing material such as carbonaceous minerals. They require pre-treatment methods such as roasting, pressure oxidation, or bioleaching (Haque 1987; Fleming 1992; Marsden and House 2006). Moreover, pre-oxidation of sulfide minerals generates sulfuric acid and ferric ions that require neutralization prior to cyanidation, which can be avoided when using alternative acidic cyanide-free leaching (Fleming 1992).

Considering the challenges of refractory gold ore processing by cyanidation and the need for a cyanide-free option, it is essential to evaluate the process performance and

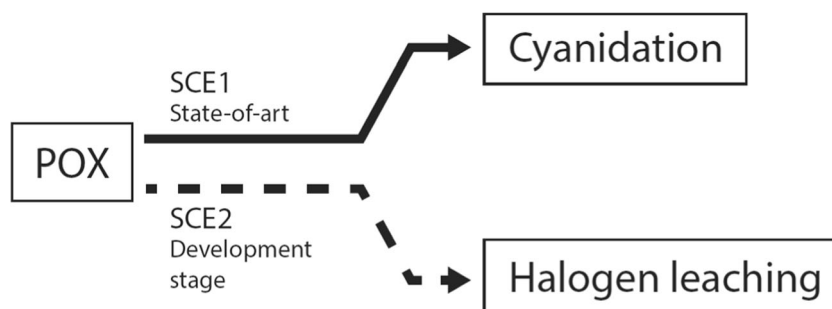
environmental impacts of cyanide-free development-stage processes prior to commercialization. The HSC Chemistry Sim module (Outotec 2018) creates metallurgical steady state simulations and mass balances, which can be utilized as life cycle inventory in GaBi LCA software (thinkstep 2019). Previously, simulation combined with life cycle assessment has been used by several researchers to evaluate different metallurgical processes (Kotiranta et al. 2015; Reuter et al. 2015; Ghodrati et al. 2017; Llamas et al. 2019). However, the compatibility of HSC Sim and GaBi has not yet been adapted to the hydrometallurgical processing of gold, in particular to the cyanide-free process options. The objective of this paper is to present an early-stage evaluation of the environmental impacts of the gold halogen process and to compare that to the state-of-art cyanidation. GaBi LCA software was used to estimate the global warming potential (GWP), acidification potential (AP), terrestrial eutrophication potential (EP), and water depletion (Water) of the process.

2 SCE1 and SCE2 processes

In the state-of-art industry, refractory gold ore is pre-treated prior to cyanidation to liberate the gold (La Brooy et al. 1994; Fraser et al. 1991). Known pretreatment methods are roasting (Hammerschmidt et al. 2016), pressure oxidation, (POX) (Mason et al. 1985; Thomas et al. 1990; Pangum and Browner 1996; Marchbank et al. 1996) and bioleaching (Iglesias and Carranza 1994; Brierley and Brierley 2001). Figure 1 presents the scenarios investigated in this study. The chosen processing route was POX of refractory concentrate and subsequent cyanidation (SCE1). This is compared to the development-stage process of POX followed by halogen leaching (SCE2). The resulting gold can then be processed through smelting to produce gold bullion. The following sections describe briefly the chemistry of the unit processes, i.e., POX, cyanidation, and halogen leaching used in SCE1 and SCE2. A more detailed description is presented in the Electronic Supplementary Material (ESM) in sections S1.1, S1.2 and S1.3.

Theoretical gold recoveries and process arrangements based on the literature were used in this study. It must be noted that the results are indicative, as the material-specific gold recoveries and the detailed method of gold recovery require tailored experimental test work for each raw material separately. The raw material used as the gold concentrate in the current study was mainly similar to that investigated by Fleming et al. (2008); therefore, similar conditions were used in the POX simulation, indicating full liberation of gold during the process. There is a significant amount of published information regarding the cyanidation of gold, whereas halogen leaching has not been reported as comprehensively. The flowsheet built for halogen leaching is based on literature data and includes

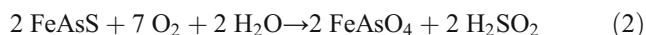
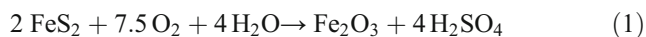
Fig. 1 The studied scenarios SCE1 consisting of POX and cyanidation (state-of-art) and SCE2 consisting of POX and halogen leaching



some modifications required for industrial scale, adopted from known technologies.

2.1 Pressure oxidation

Generally, POX targets pyrite and arsenopyrite oxidation for the complete liberation of gold (Eqs. 1 and 2). The POX is performed at a temperature between 180 and 225 °C, a total pressure of between 1900 and 3380 kPa (275 and 490 psia), and an oxygen partial pressure of at least 173 kPa (25 psia) for a period of at least 60 min (Thomas et al. 1990). Typically, the oxidation degree of sulfide sulfur aimed for is in the range of 50 to 95%. This is affected by the amount of sulfide sulfur, the nature of the sulfides in the ore, and the distribution of gold.

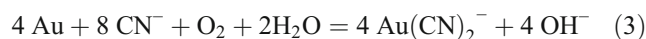


During process, it has been suggested that iron precipitates as hematite through formation of iron sulfate, which also can form jarosite precipitate. Arsenic forms a stable precipitate with iron as ferric arsenate (Haque 1987; Fleming 1992). Sulfuric acid is generated in situ in the autoclave under operating conditions, Eqs. 1 and 2, see ESM S1.1 for the reaction mechanisms. It is beneficial to maintain excess sulfuric acid in the autoclave, in order to promote oxidation and prevent preg-robbing phenomena (Marchbank et al. 1996; Fleming et al. 2008).

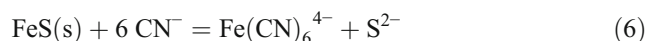
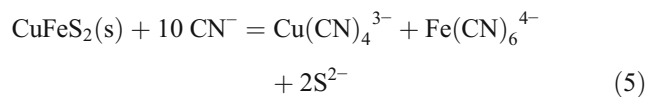
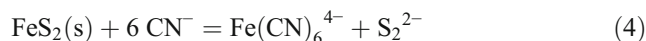
2.2 Cyanidation

Currently, cyanidation is the predominant process for gold leaching. Cyanide is introduced into the system mainly as sodium, calcium, or potassium cyanide (Marsden and House 2006). In aqueous alkaline solution, gold oxidizes and dissolves in Au(I) form and complexes with CN^- to form an $\text{Au}(\text{CN})_2^-$ complex. The stability of dicyanoaurate ion ($\log \beta_2 = 39.3$, $\beta_2 = 2 \times 10^{38}$) is significant compared to other complexes. It is known that high gold recovery can be achieved with cyanidation. Recoveries of over 90% are

reported in the literature, required for economically feasible processes. However, with direct cyanidation, refractory-type ores are defined to reach less than 80% recovery. The overall dissolution reaction of gold in aerated, alkaline cyanide solutions proposed by Elsner (1846) is shown in Eq. 3:



One of the prevailing challenges in cyanidation is related to impurities such as copper and reactive sulfides such as pyrrhotite. Copper forms stable cyanocomplexes, which increase the consumption of sodium cyanide in the process (Marsden and House 2006). Pyrrhotite and other sulfides can lead to high oxygen requirements due to the oxidation reactions of Fe^{2+} to Fe^{3+} and sulfide to sulfate (La Brooy et al. 1994). In addition, other impurities, for instance As, Ni, Sb, and Zn, can increase the consumption of either cyanide or oxygen, hindering the recovery of gold (Aylmore and Muir 2001). Furthermore, carbonaceous and clay-type material in the ore enhance the preg-robbing phenomena by absorbing Au cyanoanions and preventing the dissolution of Au (La Brooy et al. 1994). The possible cyanide-consuming dissolution reactions of sulfide minerals are suggested in Eqs. 4–6 (Zhang et al. 1997; Kianinia et al. 2018):



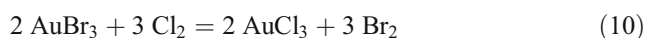
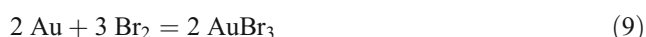
Due to the oxidizing conditions in cyanidation, the forming S^{2-} can oxidize further to sulfate (Zhang et al. 1997). These minerals should be transformed to another form prior to cyanidation, for example hematite, which is insoluble in cyanidation (Hayes 1985). Carbon in leach (CIL) has been shown to provide higher recovery of gold from refractory-type ores (Fleming 1992). Gold is stripped from the activated

carbon in the elution process to produce a smaller volume of gold-rich solution from which the gold can be recovered through electrowinning. The gold cathodes are subjected to smelting and cast into gold bullion. In order to decrease the environmental risks related to CN, the cyanide solutions are detoxified. The most common oxidation process for CN is the sulfur dioxide assisted process, where Na_2SO_3 or $\text{Na}_2\text{S}_2\text{O}_5$ is used as a source of sulfur dioxide. A mixture of sulfur dioxide and air oxidizes the free cyanide in solution according to Eq. 7 into a form in which it can be disposed of (Marsden and House 2006):

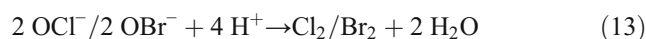
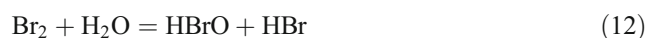


2.3 Halogen leaching

Chlorine has been known to leach gold from ores and concentrates since the 1800s (Rose 1898). A process using halogens recycled as hypochlorites for gold leaching from refractory gold ores has been patented recently (Lalancette et al. 2015a). This process was chosen for comparison as it is one of the closest to commercialization among the cyanide-free options. In the process, precious metals are dissolved by the action of halogens or halogen derivatives and recovered from the pregnant solution, while facilitating the recycling of halogens from the resulting barren solution. The redox potential of the solution has to be maintained at appropriate values, in the range of 0.7 to about 1.2 V vs. Ag/AgCl (Lalancette et al. 2015a), and more specifically 0.8 V vs. Ag/AgCl in order to dissolve gold (Marsden and House 2006). The recovery of gold can be done with activated carbon. However, in the case of halogen leaching and particularly with bromine, the halogens have a tendency to form stable halogenated compounds with carbon. This disturbs the recycling of the halogens and creates halogenated carbon disposal problem. Consequently, an alternative recovery method called silica adsorption has been proposed (Lalancette et al. 2015a). The gold silica/precipitate product can be directly subjected to a pyrometallurgical process to produce gold bullion. In the halogen leaching, halogens such as chlorine and bromine are used as free halogens for the oxidation of precious metals and are recycled by electrolysis of the barren solution in the form of hypohalides. (Lalancette et al. 2015a) According to Lemieux et al. (2014), there is interaction with elemental Cl_2 , which oxidizes the dissolved Br^- ions to Br_2 (Eq. 8), and the corresponding formation of Cl^- . Therefore, Cl_2 will generate Br_2 for efficient gold oxidation (Eq. 9). The product, AuBr_3 , will be further transformed to AuCl_3 (Eq. 10).



The Cl_2 and Br_2 gases will react with water to form hypohalides, as shown in Eqs. 11 and 12, whereas the liberation of free halogens follows Eq. 13. The recycling of halogens is based on the electrolysis in the simulated process, and the excess halogens are then scrubbed with sodium hydroxide. A more detailed process description is found in the ESM section S1.3.



3 Process simulation

The HSC Sim module was used to build the flowsheet simulations. Each simulation is based on the modeling of selected and defined unit processes and their continuum with the known and/or published chemistry. The user defines the input(s) and its/their mineralogical/chemical composition (such as Table 1) as well as possible chemical reactions and their progress. This allows the steady-state simulation of the output(s) and energy balance of the unit. The output of the first unit is the feed for the next unit(s) or product or bleeds out of the process, and thus, the entire flowsheet modeling allows the evaluation of mass and energy balances, losses, and emissions in the process accurately. The parameters of each unit are based on published literature or estimates values made by a person skilled in industrial

Table 1 The balanced mineralogy of the gold concentrate material used in the simulation of SCE1 and SCE2 (Jaatinen 2011)

Mineral	Value	Unit
Au	38.7	ppm
Ag	33	ppm
FeAsS	13.8	%
$\text{Cu}_{10}\text{Fe}_2\text{Sb}_4\text{S}_{13}$	0.34	%
C	1.95	%
FeS_2	32.1	%
Fe_2O_3	2.27	%
NiSb	0.22	%
PbS	0.06	%
Sb_2S_3	0.19	%
$\text{FeO} \cdot \text{TiO}_2$	1.71	%
$\text{NaAlSi}_3\text{O}_8$	4.82	%
$\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$	15.22	%
CaCO_3	7.01	%
MgCO_3	5.31	%
SiO_2	15.02	%
Sum	100	%

process design. In the current work, three models were built: POX, cyanidation, and halogen leaching. POX is the pretreatment step used prior to both gold cyanidation and halogen leaching. Therefore, in the current work, POX is an identical step followed by either cyanidation or halogen leaching.

3.1 Gold raw material used in simulation

The raw material used in the simulation is refractory gold concentrate, originating from a Finnish gold deposit (Jaatinen 2011). Based on the mineralogical and chemical analysis, a balanced composition of the material was calculated (Table 1). In the simulation, the dry concentrate feed to the POX is 40.2 t/h, corresponding to an annual production of approximately 10,000 kg of gold.

3.2 Pressure oxidation simulation flow sheet

The process flowsheet of POX is presented in Fig. 2 and the parameters for each unit in POX simulation in Table 2.

In the simulation, the refractory gold concentrate (40.2 t/h) is first subjected to pre-acidulation. The input material is flotation concentrate; thus, some water (23% of the feed) enters the process with it. The pre-acidulation unit introduces a significant amount of freshwater into the process in the current simulation. Slurry, with an optimal solids concentration of 12% (Conway and Gale 1990) for 30% sulfide content, is pre-heated according to the patent of Thomas et al. (1990), utilizing the excess heat of flash tanks. However, direct steam as such would challenge the water balance of the autoclave, which motivates the utilization of a mixer to provide 100 °C steam for preheating. It is likely, that the exothermic reactions in POX and the mineralogy of the studied material may enable the autoclave to maintain the required temperature without preheating. POX is conducted at 190 °C with 12% slurry, the parameters being similar to those published by Fleming et al. (2008), for a very similar raw material. Flash tanks release 29.2 and 32.6 t/h of steam for heat utilization and 384 t/h of slurry to the thickener. The underflow (UF,

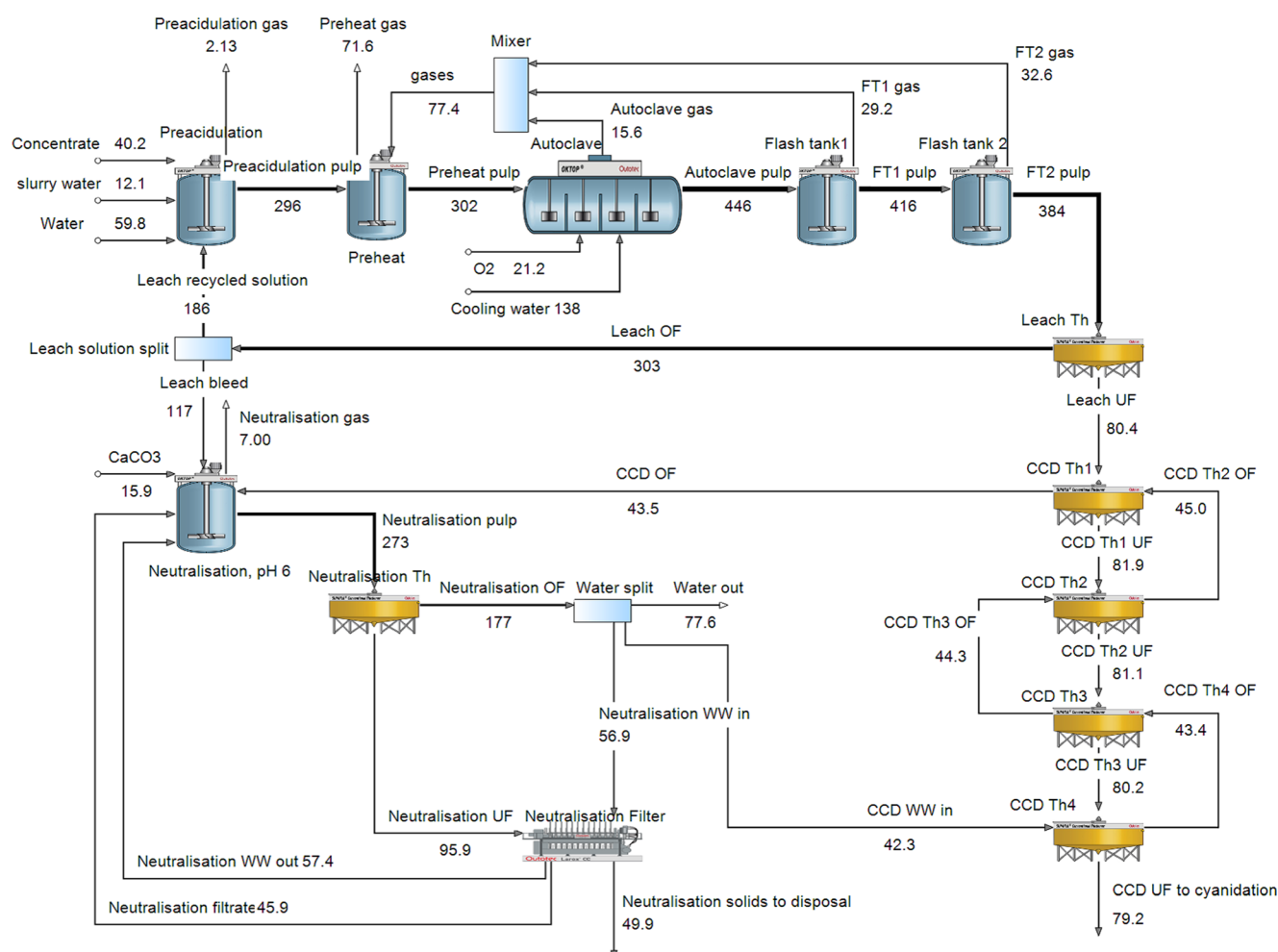


Fig. 2 The flowsheet of POX simulation for refractory gold concentrate, showing mass balance (t/h) for each flow with ca. 10,000 kg/a gold production

Table 2 The parameters used in the simulation of the POX process

Unit process	Parameter	Value	Control unit	Reference
Controlled parameters				
Preheat	Slurry concentration	12%	Pre-acidulation	Estimation based on Conway and Gale (1990)
Autoclave	Oxygen partial pressure	100 psi	Autoclave	Fleming et al. (2008)
Autoclave	Fe ³⁺ concentration	10 g/L	Autoclave	Estimation
Autoclave	As concentration	1 g/L	Autoclave	Estimation
Autoclave	Free acid concentration	85 g/L after autoclave	Leach solution split	Fleming et al. (2008)
Other parameters				
Autoclave	Temperature	190 °C		Fleming et al. (2008)
Autoclave	Sulfide mineral oxidation degree	99.8%		Fleming et al. (2008)
CCD circuit	solid conc. in underflow	40%		HSC Thickener standard
Filtration	Filtration efficiency	96%		HSC Filter standard
Filtration	Filtration cake moisture	25%		HSC Filter standard
Filtration	Washing water amount	1.5 m ³ /t of solids		HSC Filter standard
Neutralization	pH	6		Estimation

80.4 t/h) is subjected to counter-current decantation (CCD) for washing of solids, whereas the overflow (OF) is partially (186 t/h) circulated back to pre-acidulation. The rest of the OF solution (117 t/h) is routed to neutralization using CaCO₃ (15.9 t/h). The neutralized solution is then used in the washing of neutralization residue and leach residue in the CCD circuit. The excess water (77.6 t/h) must be bled out of the process. This water is neutralized at pH 6 but contains sulfates (17.6 g/L). It can be used in subsequent unit processes or disposed of after water purification, as one recommended sulfate limit being 1000 mg/L in mine waters in Europe (Bowell 2004). The process water removed in the neutralization, as well as the evaporated water of flash tanks, could be partially reused to replace the amount of used fresh water in pre-acidulation. However, the current model is based on the use of fresh water in the pre-acidulation.

The main principles and observations of the simulated POX process are described in the following sections.

3.2.1 Sulfuric acid consumption

Several patents suggest sulfur acid concentration of preferably less than 25 g/L should be maintained by sulfur oxidation in POX (Thomas et al. 1990; Marchbank et al. 1996). However, the results of the current study suggest that up to as much as 85 g/L of sulfuric acid can be maintained without addition of sulfuric acid chemical. The current simulation is operated at high acidity as it has been suggested to prevent preg-robbing and improve

gold recovery by 2–3%. In addition, this may allow more consistent and stable metallurgical performance for highly refractory gold concentrate (Fleming et al. 2008). The leach solution is reused in the pre-acidulation unit.

3.2.2 Neutralization

The simulation reveals that the calcium carbonate consumption in solution neutralization is 398 kg/t concentrate, based on stoichiometric reaction. The neutralization products are jarosites (Pangum and Browner 1996) and oxides and hydroxides such as FeAsO₄, Cu (OH)₂, CaSO₄·2H₂O, and Al (OH)₃. In addition, the simulation shows that some of the dissolved arsenic (1 g/L, AsO₄[−]) will end up in the neutralization stage and may consequently re-dissolve from the solid tailings. For this reason, the resulting tailings are characterized as hazardous substances and must be discharged into specific ponds. Contemporary practices for this kind of tailing materials are paste thickening and use as a backfill in underground mines, for example (Pöyry 2015).

3.2.3 Off gases

The results confirm that carbon dioxide is produced during POX process. In the simulations, these gases are not treated and are released into the environment as such. The effect of the CO₂ gases produced is seen in the life cycle assessment section.

3.2.4 Treated concentrate

POX produces an oxidized concentrate, subjected to a gold leaching process after CCD washing, in the form of thickener underflow. The solid concentration of the slurry is 36%, it is acidic, and it contains 28.6 t/h of undissolved silicate minerals. Sulfide mineral dissolution in POX has liberated the gold, whose concentration has increased to 54.3 ppm in underflow slurry.

3.3 State-of-art cyanidation flow sheet

In the SCE 1 process, cyanidation is conducted for POX-treated gold concentrate. The unit processes are modeled as one unit; however, in a real process, the carbon-in-leach/CIL circuit consists of multiple reactors. The oxidized concentrate, i.e., the UF of CCD circuit (79.2 t/h), is subjected to pH control by $\text{Ca}(\text{OH})_2$ (0.88 t/h). Due to the risk of the formation of lethal hydrogen cyanide gas, maintaining a high pH is crucial for safe plant operation. 79.2 t/h of slurry is subjected for CIL, where gold dissolves and adsorbs on carbon, at pH 10 and keeping the NaCN level at 0.5 g/L. After leaching, the bleed solution is routed to the cyanide destruction unit, which

consumes air (65 kg/h), Na_2SO_3 (83 kg/h, calculated from the composition of the flow) and $\text{Ca}(\text{OH})_2$ (49 kg/h), after which the tailings can be discharged into ponds.

In a real process, the loaded carbon is gathered in hoppers prior to proceeding to the next unit process in order to adjust the batch sizes. In the steady-state simulation, these hoppers were excluded. However, the recovery process was simulated as a 1-t batch, for equipment sizing purposes.

Of loaded carbon, 1 t/h is treated in the recovery process, where the carbon is first washed with 3% HCl solution to remove impurities. In the elution column, gold is stripped from the carbon by soaking in 35 g/L NaCN (0.46 t/h) and 12.5 g/L NaOH (0.17 t/h) solution, and then washed with water (12.2 t/h), which is five times the carbon bed volume. Subsequently, gold is electrowon from the solution and a similar detoxification is used for the cyanide solution, consuming 1.3 t/h of Na_2SO_3 , 1.15 t/h of air and 0.79 t/h of $\text{Ca}(\text{OH})_2$.

The results of the mass balance for each unit process flowsheet of gold leaching by cyanidation are shown in Figs. 3 and 4. The loaded carbon output of leaching was increased to and input of 1.08 t/h in the recovery simulation.

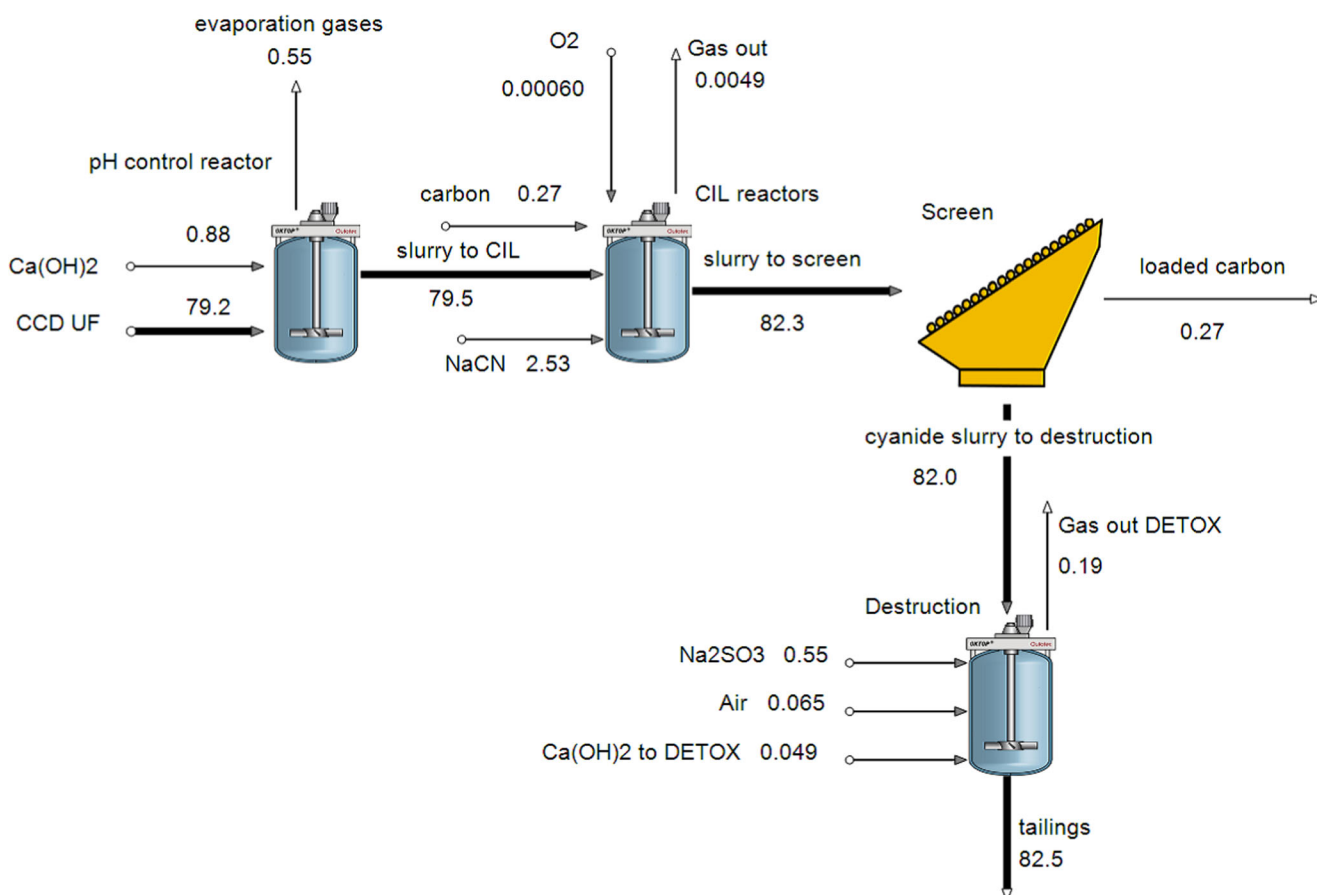


Fig. 3 The cyanidation simulation flowsheet for pressure oxidized refractory gold concentrate, showing mass balance (t/h) for each flow with ca. 10,000 kg/a gold production

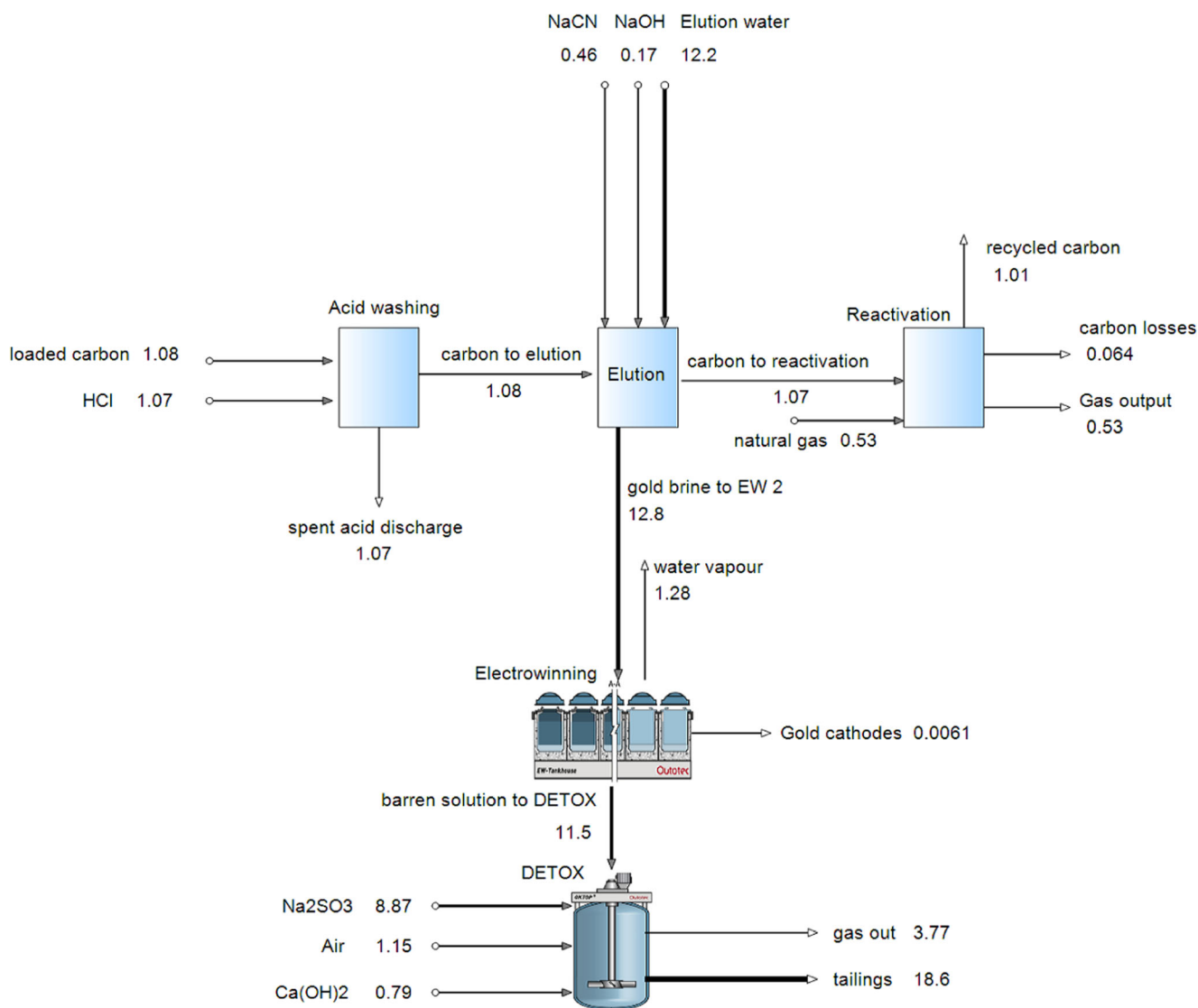


Fig. 4 The flowsheet of gold recovery from activated carbon originating from cyanidation, showing mass balance (t/h) for each flow with ca. 10,000 kg/a gold production

The parameters used in the simulation are shown in Table 3. Some of the parameters are used to control concentrations in the reactors, etc. Other parameters are not effectively used in the simulation but are compared with the results obtained from the simulation.

The main principles and observations of the simulated cyanidation are described in the following sections.

3.3.1 Gold recovery

The simulated gold recovery from the feed material was found to be 98.5%. This is based on 99.8% leaching and 100% adsorption into carbon during instant recovery by CIL. This instant recovery on carbon allows minimization of gold losses into (i) the leach residue or (ii) the washing waters of the leach

residue. In fact, no washing of the leach residue is conducted in cyanidation; the leach residue is discharged into a tailings pond after detoxification, from where the solution can be partially recycled back to the process.

3.3.2 Cyanide consumption

In the current simulation, the cyanide consumption in the CIL unit was found to be 0.98 kg/t of concentrate. This is higher than typical cyanide consumptions (0.25 to 0.75 kg/t) for free-milling gold ores (Marsden and House 2006). However, when the feed material contains significant amounts of cyanide consumers and/or high silver content (i.e., 20 g/t), cyanide consumption may increase up to 1 to 2 kg/t (Marsden and House 2006).

Table 3 The parameters used in the simulation of the cyanidation process

Unit process	Parameter	Value	Control unit	Reference
Controlled parameters				
CIL reactors	NaCN concentration	0.5 g/L	CIL reactors	Marsden and House (2006)
CIL reactors	Solids concentration	35%	CIL reactors	Marsden and House (2006)
CIL reactors	Activated carbon	5 g/L	CIL reactors	Marsden and House (2006)
CIL reactors	Gold loading	5–10 kg Au/t carbon	CIL reactors	Marsden and House (2006)
Cyanide destruction	SO ₂ [−] consumption	3–4 kg per 1 kg free cyanide	Destruction	Marsden and House (2006)
Acid washing	HCl concentration	1–5%	Acid washing	Marsden and House (2006)
Acid washing	Acid consumption	25–50 kg/t carbon	Acid washing	Mular et al. (2002)
Elution AARL	NaCN concentration	20–50 g/L	Elution	Marsden and House (2006)
Elution AARL	NaOH concentration	10–15 g/L	Elution	Marsden and House (2006)
Other parameters				
pH control reactor	pH	10.5–11		Marsden and House (2006)
CIL reactors	Temperature	25 °C		Marsden and House (2006)
CIL reactors	Lime consumption	13 kg/t solids		Fleming et al. (2008)
CIL reactors	NaCN consumption	1 kg/t solids		Fleming et al. (2008), Marsden and House (2006)
CIL reactors	Activated carbon consumption	20–40 g/t ore		Marsden and House (2006)
Cyanide destruction	pH	9		Marsden and House (2006)
Cyanide destruction	CuSO ₄ catalyst	30–90 Cu ²⁺ g/t		Marsden and House (2006)
Acid washing	Temperature	80 °C		Marsden and House (2006)
Elution AARL	Temperature	110–120 °C		Marsden and House (2006)
Elution AARL	Pressure	170–200 kPa		Marsden and House (2006)
Reactivation	Temperature	750 °C		Marsden and House (2006)
Gold EW	Au concentration	10 g/L		Marsden and House (2006)
Gold EW	NaCN concentration	2%		Marsden and House (2006)
Gold EW	NaOH concentration	0.5%		Marsden and House (2006)
Gold EW	Temperature	80 °C		Marsden and House (2006)

3.3.3 Lime consumption

Lime, Ca (OH)₂, is the preferred chemical for pH control in cyanidation. Typical concentrations of 0.15–0.25 g/L of calcium hydroxide are used, corresponding to lime consumptions of 0.15–0.5 kg/t of ore (Marsden and House 2006). However, these consumption values are for non-acidic- or non-alkali-consuming ores. In the studied system, POX was run at high

acidity (see Sect. 3.1) to support higher gold extraction from preg-robbing raw material—therefore also notably higher lime consumption was observed (30.6 kg/t of concentrate). This is exceptionally high level; however, as high as 13 kg/t has also been reported by Fleming et al. (2008). In the end, the consumption of Ca (OH)₂ needs to be optimized vs. gold value; however, in the current study, the high gold extraction was emphasized.

disposed of. The chloride solution (12.5 t/h) is recycled back to leaching through hypohalide electrolysis.

The mass balance for each unit in halogen leaching flowsheet (SCE2) is shown in Fig. 5. The process parameters used in the simulation are listed in Table 4. Parameters used in simulation control are shown separately from other controls. Filter and thickener controls are standard controls for internal models found in HSC Sim equipment models.

The main principles and observations of the simulated halogen leaching are described in the following sections.

3.4.1 Gold recovery

In the SCE2 process, the gold extraction values are based on Lalancette et al. (2015a), Lalancette et al. (2015b), and Lemieux et al. (2014), reported to be from 95% to as high as 98% for 2-h leaching. For refractory-type material, longer leaching times can be required, thus also expecting higher gold extraction. Different to cyanidation, the gold halide leaching in SCE2 does not have in situ gold recovery, but gold recovery is conducted after S/L separation by adsorption to silica, with reported recovery of as high as 99.98% (Lalancette et al. 2015b). According to these values, the simulation predicts 87.3% gold recovery. The low total Au recovery is explained by the following facts:

- In the studied case, in total, 1% of gold is lost to leach residue.

- There is no instant gold recovery in the halide process. This causes challenge as the gold present in the PLS also ends up to the moisture of the leach residue. The amount of washing water in filtration is limited (1.5 m³/t solids) to keep the process water balance. Therefore, minor amount of PLS, and gold, is always present in the residue.
- Also, minor amount gold does not dissolve from the raw material.
- The used washing water (1.5 m³/t solids) displaces some of the gold containing PLS at the filter; this brings along 11% of soluble gold with it to the evaporation stage. In SCE2, the latter is partly lost due to precipitation to leaching residue that can be prevented by washing the residue first with halide containing washing solution.

3.4.2 Process design aspects in halogen leaching

In the halogen process, halogen gases are produced in situ by redox control. This makes it possible to produce only the required amount of gases and circulate the halogens as hypohalides. The flowsheet was developed according to the literature; however, some modifications were made. Some of the gold-depleted solution was used to maintain the solids concentration in the leaching in order to recycle chlorides as early as this stage. Due to water

Table 4 Parameters used in simulation of the halogen leaching process

Unit process	Parameter	Value	Control unit	Reference
Controlled parameters				
Leaching	NaCl concentration	60 g/L	Leaching	Lalancette et al. (2015a)
Leaching	NaBr concentration	2 g/L	Leaching	Lalancette et al. (2015a)
Leaching	Solids concentration	20%	Split 1	Lalancette et al. (2015a)
Silica adsorption	Gold loading	30%	Silica adsorption	Lalancette et al. (2015a)
Other parameters				
Leaching	pH	1.5		Lalancette et al. (2015b)
Leaching	Temperature	40–50 °C		Lalancette et al. (2015a)
Filtration	Filtration efficiency	96%		HSC Filter standard
Filtration	Filtration cake moisture	25%		HSC Filter standard
Filtration	Washing water amount	1.5 m ³ /t of solids		HSC Filter standard
Thickener	Solid conc. in underflow	40%		HSC Thickener standard
Silica absorption	pH	1.5		Lalancette et al. (2015a)
Neutralization	pH	6		Estimation

accumulation, an evaporation step was added to produce high-chloride brine, which could possibly be used in leaching.

3.4.3 Leach residue washing—water balance—environmental regulations for LR

Due to challenges observed in S/L separation in halide process, different options for leach residue washing were simulated. The presented flowsheet shows the option, where accumulating water is evaporated in order to achieve concentrated brine, which could be recycled back to leaching.

Three other options were investigated for leach residue (LR) washing, a schematic flowsheet presented in Fig. 6:

- Case 1: LR is not washed.
- Case 2: LR is washed with water.
- Case 3: LR is washed with PLS after gold recovery.

In all cases, 1% of the gold left in residues is lost because this gold is not leached in the leaching stage. In addition, part of the leached gold in case 1 (0.02 g/L) is lost with solids as moisture, due to the fact that the washing liquids are not circulated back to the leaching reactors, as in cases 2 and 3. However, by not washing the residues and circulating these waters to the process, the accumulation of waters can be avoided. In case 2, accumulation of water in the process was observed. Around 92.2 t/h of solution needs to be taken out from the process. This solution has a chloride concentration of 35.4 g/L, which means that further treatment is necessary before the solution can be either used as process water or disposed of. In addition, the chemical consumption of NaBr and NaCl is higher along the chemicals bled out of the process. In case 3, the leach residue is washed with filtrate of Au/SiO₂ product

filter. In this case too, water accumulation is avoided to some degree. However, some gold is lost to the residues. These alternatives concluded that it would be best to gather washing waters to evaporation, in order to produce a concentrated brine, creating a possibility to recycle chloride chemicals.

3.4.4 Neutralization residue—water treatment

The neutralization residue is washed, and the filtrate is circulated back to the process. The moisture of the neutralization cake is 25%, mostly water as the washing efficiency is 96%. The chloride concentration in the presented flowsheet is 1.38 g/L in the solid residue flow and the washing waters have 7.44 g/L of Cl[−]. Therefore, in addition to leach residue, the process produces a waste fraction with ca. 345 ppm of Cl[−], which is well below environmental regulations of landfill qualifications. The chloride concentration limits for conventional landfill are 15,000 ppm and for hazardous landfill 25,000 ppm in Finland (Wahlström et al. 2006).

3.4.5 Tank house operation

The water balance also affects the operation of hypochlorite electrolysis. The electrolysis of hypochlorite can be energy-consuming. Therefore, the more process liquids are subjected to electrolysis, the more energy is consumed in the process. The actual patent describing the halogen process does not describe the parameters used for hypohalide electrolysis; thus, it was assumed to be a similar process to other hypohalide electrolysis processes presented in the literature.

According to the Childers et al. (2007) patent for producing hypochlorite by electrolysis, the electric current is controlled to keep power consumption below 2.5 kWh/pound of hypochlorite produced and 2.5 kg of NaCl is consumed to produce

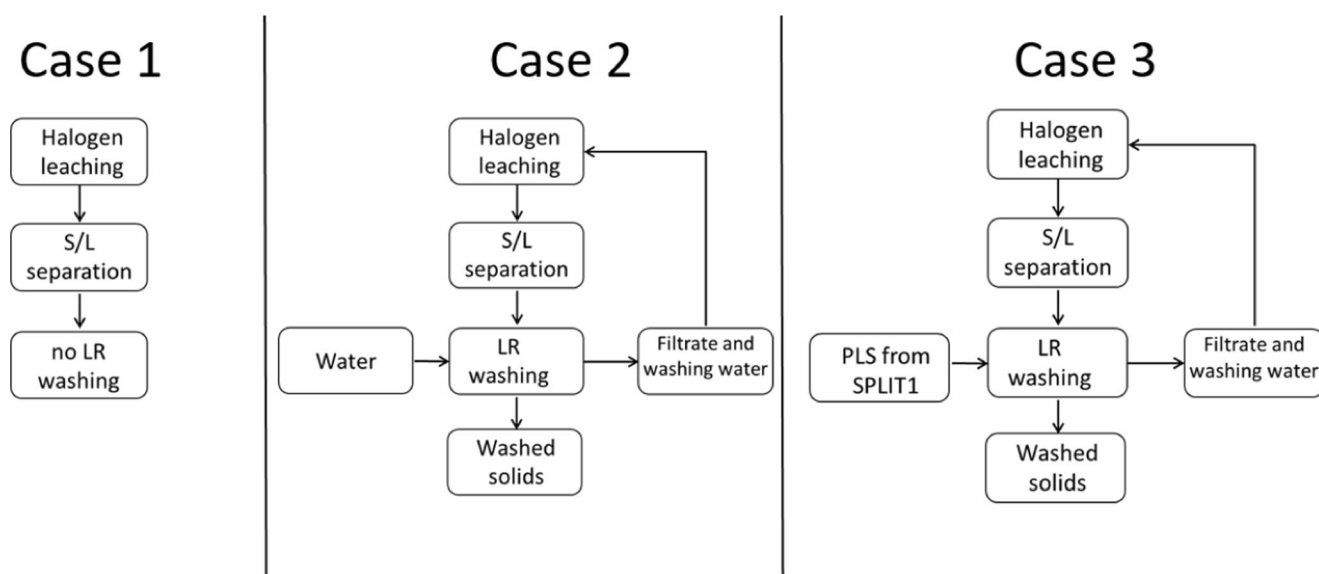


Fig. 6 The schematics of cases investigated to avoid water accumulation in the halogen process

1 kg NaOCl. Bashtan et al. (1999) report consumption of 2.4 kg NaCl to produce 1 kg of NaOCl and power consumption of 3.1 kWh/kg NaOCl produced. In addition, Kuhn and Lartey (1975) reported the NaCl consumption of 3–3.5/kg NaOCl produced and power consumption of 4.4–8.8 kWh/kg NaOCl. An average value of 3.1, 4.4, and 5.5 kWh/kg NaOCl, corresponding to 16 MJ/kg NaOCl, was used to estimate the power consumption of electrolysis.

3.4.6 Chemical consumption

The NaBr concentration in leaching should be 2 g/L, and the reported Cl^- concentration in PLS is 32.32 g/L (Lalancette et al. 2015a), which corresponds to a NaCl concentration of 60 g/L. These concentration values were used in the simulation to control the inputs of NaBr and NaCl chemicals, and thus in the presented flowsheet, the NaBr consumption is predicted to be 0.14 t/h and NaCl consumption 4 t/h. These values correspond to 5 kg NaBr/t and 140 kg NaCl/t concentrate. These levels of consumptions are significantly higher compared to cyanidation. However, electrolysis provides a method to partially recycle the chlorides. Moreover, due to lack of published data, the solid concentration in leaching is given as 20%, which corresponds to higher solution volumes in leaching and thus higher chemical consumption.

3.4.7 Second gold recovery process

As the total gold recovery in halogen leaching was notably low, the addition of a second gold silica adsorption step, described herein as SCE2b, was simulated for the recovery of gold from wash water (WW4) assuming no gold precipitation during leach residue washing. In this way, a higher total Au recovery of 98.5% could be predicted by the simulation. However, the use of a second recovery unit has not been reported in the literature.

3.5 Comparison of mass (and energy balance) in SCE 1 and SCE2

The values presented in the investigated scenarios are for a throughput of 40.2 t of concentrate. This corresponds to 28.6 t of dry concentrate after POX, routed to gold leaching as 36% slurry. It can be seen that, in general, the consumables in halogen leaching were higher compared to cyanide, see Table 5. However, the lime consumption is higher in cyanidation due to the alkaline nature of the cyanidation process whereas, in halogen leaching, lime is used only for neutralizing the solutions. The high water consumption in halogen leaching originates from the need to wash chloride out of the leach residue and neutralization solids. The residues were washed in filters using 1.5 m³ washing water/t of solids, with 96% efficiency.

The electricity consumption was evaluated based on the simulation model by taking into account the most electric energy-consuming unit processes. The equations and calculation methodology are presented in detail in the ESM S1.4. The higher electricity consumption in halogen leaching (SCE2) compared to cyanidation (SCE 1) can be explained by the hypohalide electrolysis (ESM S1.4), being an average of 4.3 kWh per kg of hypochlorite produced.

The observations here highlight some of the challenges related to simulation of development-stage cyanide-free processes—although the evaluation of these processes is necessary prior to upscaling to industrial scale, there is only limited information compared to proven technologies (the process flow sheets have not been published or designed in detail), which may lead to large variation in the estimation of the final mass and energy balances and furthermore in the life cycle assessment. It is evident that the halogen process (SCE2) suffers from handling the washing waters and gold lost to residues or sidestreams, as gold leaching and recovery are conducted in separate stages. The benefit of cyanidation (SCE1) is the simultaneous recovery of gold on carbon by the CIL leaching method, creating a smaller volume of gold rich solution in the elution process as well as production of detoxified residues without washing requirements.

4 Life cycle assessment

4.1 Assumptions

Life cycle assessment (LCA) can be used to evaluate the environmental impact of a product, a process, or an activity during its lifetime from raw material to end-of-life. LCA is essentially an environmental tool, which can be adapted to evaluate the environmental impacts of metallurgical processes. ISO 14040 and ISO 14044 standards describe the methodology of LCA (ISO 14040 2006; ISO 14044 2006). The use of LCA methodology is increasingly used to assess the environmental sustainability of metallurgical processes (Norgate et al. 2007; Norgate and Jahanshahi 2010), however, few studies about gold processes have been presented (Norgate and Haque 2012). The method used in this study, combining HSC Sim and GaBi software, has been adopted by Llamas et al. (2019), Ghodrati et al. (2017), Reuter et al. (2015) and Kotiranta et al. (2015). However, the methodology has not yet been excessively used to evaluate hydrometallurgical gold processing.

4.1.1 LCA assumptions

In this study, LCA has been performed on two simulation models, one consisting of POX and cyanidation (SCE1), and the other POX and halogen leaching (SCE 2), both utilizing a refractory gold concentrate as the raw material. The

Table 5 Comparison of input quantities to treat 40.2 t of concentrate in SCE1 and SCE2

Input	SCE1: POX and cyanidation	SCE2:POX and halogen leaching	Unit
Dry concentrate	40.2 POX/28.6 leaching	40.2 POX/28.6 leaching	t
Electricity	25,277	30,579	MJ
Ca(OH) ₂	17.8	16.7	t
Air	0.5		t
Oxygen	21.2	25.2	t
NaCN	0.1		t
NaCl		4.0	t
NaBr		0.1	t
Water	268.9	309.7	t

environmental indicators for each scenario are based on the mass balance produced with HSC Sim steady-state simulation (Chapter 3 and ESM S1.1.–S1.4.). The simulation produces a consistent life cycle inventory (LCI) (Llamas et al. 2019). HSC Sim is compatible with the GaBi LCA software, where LCI data from HSC-Sim is directly exported to. In GaBi software, the environmental indicators of global warming potential (GWP), acidification potential (AP), terrestrial eutrophication potential (EP), and water depletion (Water) are estimated. The LCI for both scenarios reference Finnish or European data in GaBi database.

4.1.2 Functional unit

In cyanidation scenario, gold is produced as cathode gold and in the halogen leaching as a silica adsorption product. The functional unit selected was 1 kg of produced gold. Both scenarios have the same input for POX and the leaching stage.

4.1.3 System boundaries

In this study, the system boundaries cover the primary raw materials and energy entering the POX and leaching unit processes up to when the product leaves the process as product, emission, or by-product as a gate-to-gate model of the refractory gold concentrate hydrometallurgical processing. The treatment and final disposing of waste streams is not included in the metallurgical simulation of the processes; thus, they are out of the system boundary of this study. However, the produced waste streams are discussed from environmental point of view. The processing route of gold and the boundaries for the LCA study are presented in Fig. 7. The geographical scope of the study is regional. The results of this study are indicative, as the scales, conditions, technologies, and operation of the modeled processes are based on literature information, not on industrial processes.

The HSC-Sim can be used for the normalization of flows. As mentioned, the flows were normalized to the production of 1 kg Au, on which the environmental indicators are based on.

The description of the LCI and the comparison of simulation model LCAs are discussed in the following sections.

4.2 Inventory for LCA

The inputs and outputs of the HSC Sim simulation model can be linked to corresponding flows in the GaBi database through the LCA Evaluation tool in HSC Sim. Table 6 lists the inventory data used for the LCA study of the simulations normalized to 1 kg of Au. The data was produced with HSC-Sim software, where the simulation models were based on data collected from the literature described in Table 2, Table 3, and Table 4. The electricity consumption in the halide process is higher due to hypohalide electrolysis, as stated previously. It can be seen that the halogen process inventory is scaled larger compared to cyanidation due to lower recoveries of gold; thus, more concentrate is needed from POX operation to leaching stage in order to produce 1 kg of gold. As a result, the environmental impact of gold leaching is greater in general. Nevertheless, the emissions are of the same magnitude in both processes. The improved gold recovery by addition of a second gold recovery unit (SCE2b) brings the halogen process more close to values of cyanidation (SCE1). The energy of cyanidation recovery process refers to the heating of the activated carbon reactivation kiln. In halogen leaching, the unspecified energy refers to the heat required mainly by not only the water evaporation unit but also in electrowinning and leaching. However, this flow was cutoff due to uncertainty of upstream process for the flow.

4.3 Life cycle impact assessment

The environmental impact indicators chosen for discussion in this study are presented in Fig. 8 for the SCE1, SCE2, and SCE2b processes.

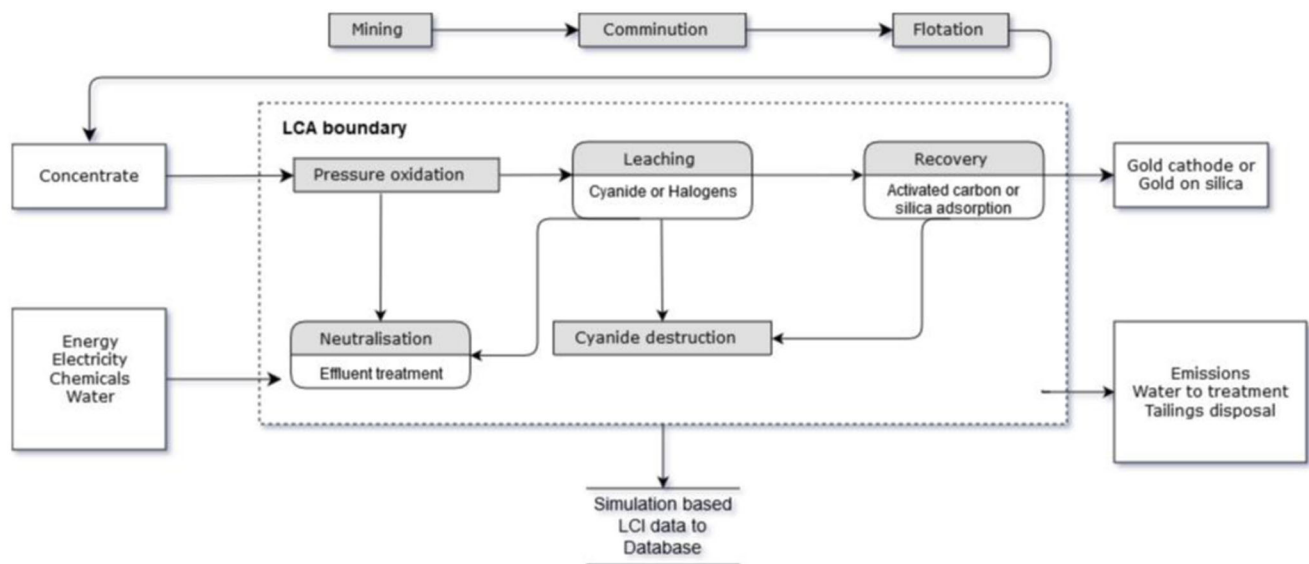


Fig. 7 The process route of gold from mining to product. The dashed line emphasizes the system boundary of the LCA conducted in this study. The simulated unit processes provide the LCI data

4.3.1 GWP

The GWP obtained in this study is 12.6 t CO₂-e/kg Au for SCE2 and 10.3 t CO₂-e/kg Au for SCE1 (Fig. 8a). The GWP estimated in halogen leaching is larger due to the lower recovery of gold (87.3%), indicating that more concentrate needs to be processed in order to produce 1 kg of gold. Thus, the impact of POX increases the GWP. In the scenario with an additional gold recovery unit (SCE2b), the GWP is lower, at 11.5 t CO₂-e/kg Au, decreasing the difference in GWP between state-of-art cyanidation and development-stage halogen leaching from 2.3 t CO₂-e/kg Au to 1.2 t CO₂-e/kg Au. The difference in GWP is greatly affected by the differences in electricity consumption between SCE1 and SCE2, as hypohalide electrolysis was suggested to be energy-consuming. Also, it needs to be noted that NaCl is included in the halide leaching LCA, but input data for NaCN was not found in the database. However, the results here indicate that the difference between a development-stage halide process (SCE2) and state-of-art cyanidation (SCE1) can be decreased through the optimization and process development, to make the halide process competitive in terms of the carbon emissions.

4.3.2 AP

The inputs affecting acidification potential are the same in both SCE1 and SCE2, as are seen in Fig. 8b. However, in halogen leaching, the production of sodium chloride increases the AP. The simulation did not predict the output gas of SO_x from the cyanide detoxification units, where sulfur dioxide is used as the reagent. Sulfur gases can be

released from detoxification units, which might increase the acidification potential of cyanidation. In addition, the NaCN was not found in database and could have impact on SCE1 acidification potential.

4.3.3 Terrestrial EP

The eutrophication potential was found to be 1.7 times higher in halogen leaching compared to cyanidation, as shown in Fig. 8c, and this is affected by the NO_x emissions. The EP of halide leaching is mostly due to NaCl production, which contributes 27.4% to the halogen leaching EP. Electricity consumption contributes 33.8% of the total eutrophication potential in cyanidation and 33.9% in halogen leaching. The EP can be lowered through efficient recycling of chloride chemicals in the plant.

4.3.4 Water depletion (water)

Water depletion describes the amount of water resources that is reduced. Water depletion was suggested to be higher in halogen leaching: 99.3 kg of water for halogen leaching compared with 77 kg of water for cyanidation, as shown in Fig. 8d. The difference is explained by the larger amount of process water consumed in halogen leaching due to the low solid concentration reported for leaching. In the cyanidation process, in contrast, the activated carbon method scales down the recovery process compared to halogen leaching, where large amounts of solution is directed to silica adsorption. The usage of fresh water can be lowered by providing a water purification plant and recovering the steam condensate from POX plant.

Table 6 LCI produced with HSC-Sim simulation normalized to 1 kg of gold

Input	Amount	Unit	Cutoff flow	Output	Amount	Unit
POX, SCE1, and SCE2						
Electricity (electric power)	3035.3	kWh		Concentrate	17,875	kg
Oxygen	13,250	kg		Carbon dioxide	5843.8	kg
Limestone	9937.5	kg		Nitrogen	131	kg
Process water	131,187.5	kg		Oxygen	2625	kg
Concentrate	25,125	kg		Process water (in slurry)	31,562.5	kg
				Solid waste	23,375	kg
				Water vapor	41,875	kg
				Wastewater	56,312	kg
Cyanidation, SCE1						
Air	41.3	kg	*	Loaded carbon	168.8	kg
Activated carbon	168.8	kg	*	Nitrogen	32.5	kg
Calcium hydroxide	550	kg		Solid waste	17,875	kg
Concentrate	17,893.8	kg		Water vapor	434.3	kg
Electricity	1273.6	kWh		Wastewater	33,687.5	kg
Oxygen	0.4	kg				
Sodium cyanide	17.5	kg	*			
Sodium sulfite	51.9	kg	*			
Water	33,406.3	kg				
Recovery, SCE1						
Activated carbon	177	kg	*	Gold	1	kg
Air	271.8	kg	*	Activated carbon [for reuse]	165.6	kg
Calcium hydroxide	129.5	kg		Carbon dioxide	12.8	kg
Electricity	83.4	kWh		Hydrochloric acid [for recovery]	4.9	kg
Thermal energy (heating)	172.4	MJ		Nitrogen	214.8	kg
Fuel, natural gas in Finland	86.9	kg		Wastewater	3219.7	kg
Hydrochloric acid	5.2	kg		Water vapor	687.9	kg
Process water	3513.1	kg				
Sodium cyanide	75.4	kg	*			
Sodium hydroxide	27.9	kg				
Sodium sulfite	218	kg	*			
Halogen leaching, SCE2						
Electricity	2598.4	kWh		Gold	1	kg
Unspecified energy (heating)	130	GJ	*	Brine	20,571.4	kg
Oxygen	37.9	kg		Hydrogen (Electrolytic steam)	12.1	kg
Calcium hydroxide	571	kg		Silica sand	3.4	kg
Concentrate	20,428.6	kg		Sulfur dioxide	0.002	kg
Silica sand	2.3	kg		Solid waste	22,121.4	kg
Sodium bromide	100	kg		Wastewater	7371.4	kg
Sodium chloride	2857.1	kg		Water vapor	45,428.6	kg
Sodium sulfite	300	kg	* remarks the cut-off flow			
Process water	71,104.9	kg				

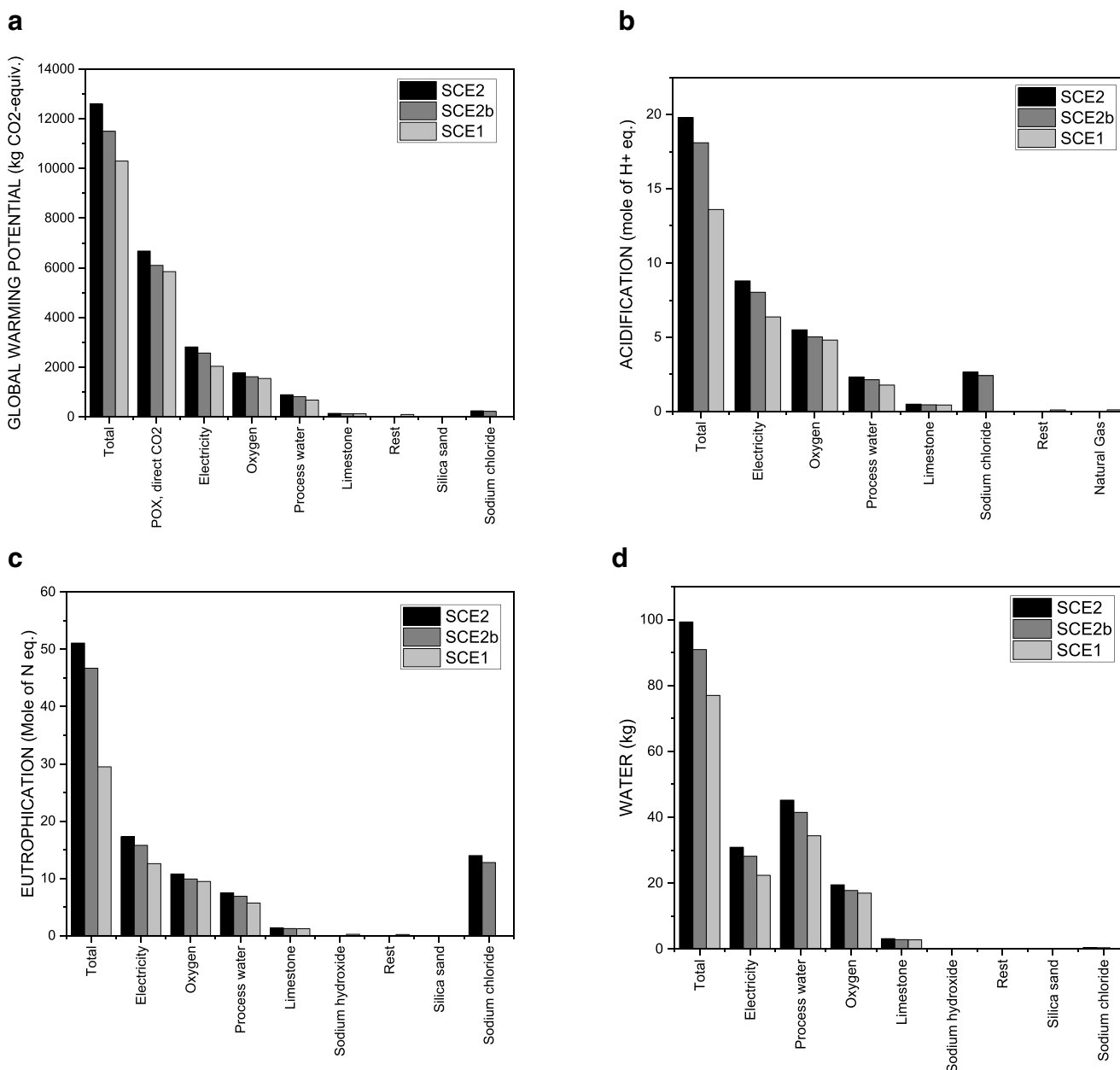


Fig. 8 Environmental impact indicators of POX followed by cyanidation or halogen leaching

4.3.5 Environmental implications of process design

The possible waste streams of hydrometallurgical processing are the solid wastes, gas discharges, and wastewaters. The aim of the processing is to produce; stable solid wastes that can be deposited safely, clean gases that can be released to the environment and purified wastewaters, which have acceptable concentrations of hazardous elements.

The composition of produced solid waste is affected by the mineralogy of the concentrate through unleached oxide and silica minerals and the produced neutralization precipitates. In this study, the concentrate includes a significant amount of arsenopyrite, which is oxidized for gold liberation. In the

autoclave conditions, most of the AsO_4^{3-} will precipitate as FeAsO_4 (scorodite) and the rest of the arsenic will be precipitated in the neutralization stage as scorodite. The solubility of scorodite has been found to be low, less than 5 mg As/L; therefore, scorodite is considered to be the most stable arsenate compound suitable for arsenic disposal purposes. (Swash and Monhemius 1998) Most of the scorodite ends up into the leach residue and will be deposited into a tailings pond.

The other dissolved impurities during pressure oxidation are precipitated as hydroxides in the neutralization unit process. These include $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, $\text{Cu}(\text{OH})_2$, $\text{Ni}(\text{OH})_2$, $\text{Al}(\text{OH})_3$ and jarosites such as $\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$

$\text{NaFe}_3(\text{SO}_4)_2(\text{OH})_6$ and $(\text{H}_3\text{O})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6$. These are also deposited into tailings pond.

The gold concentrate also contains PbS (Galena), $\text{Cu}_{10}\text{Fe}_2\text{Sb}_4\text{S}_{13}$ (Tetrahedrite), and NiSb (Breithauptite), which include lead and antimony. In sulfuric acid, pressure oxidation lead precipitates as lead sulfate (PbSO_4). This precipitate will end up in the subsequent leaching step, i.e., cyanidation or halogen leaching, indicating that the pressure oxidation wastewaters or solids will not contain lead. Antimony minerals will also leach in the pressure oxidation and precipitate as FeSbO_4 (Tripuhyite). Thus, the precipitate products (FeAsO_4 , FeSbO_4 and PbSO_4) originating from pressure oxidation will end up into the leach residue, i.e., cyanidation tailings or halogen leaching solid waste after leaching residue washing. According to Multani et al. (2016), tripuhyite is the optimal form for antimony immobilization. Lead sulfate is known to be poorly soluble to water; however, slow release of lead to environment over a long period of time can be possible.

The main gaseous emissions predicted by the simulation and shown in the GWP estimation is the CO_2 produced in the process. The concentrate includes carbonate minerals such as MgCO_3 and CaCO_3 , which produce CO_2 when leached. Many unit processes also use oxygen and air feed, resulting in gaseous emissions as the undissolved gases are released. Sodium sulfite is used for the oxidation of CN^- ion in cyanidation or reduction of gold on silica in halogen leaching. This chemical produces excess SO_2 gas; thus, minor SO_2 emissions are possible.

Mining waters have strict environmental limitations of dissolved elements in the discharged solutions. Sulfate is one of the species found in mine waters and is particularly problematic when in the form of a highly soluble sodium sulfate, as sulfates in water bodies can increase the salinity.

The wastewaters produced during the pressure oxidation do not contain notable amounts of dissolved metallic impurities after neutralization. However, as stated before, the sulfate concentration of the pressure oxidation waters (17.6 g/L) is higher than the European reference effluent value of 1000 mg/L (Bowell 2004). The geographical mine locations as well as local regulations define the finally the annual amounts of acceptable sulfate disposal. As an example, the acceptable sulfate concentration in a Finnish gold plant has been over 8000 mg/L in discharge water in the past, which has been lowered to 2000 mg/L with addition of sulfate removal process. (Pöyry, 2016) Furthermore, due to MgCO_3 dissolution, there are Mg^{2+} ions (4.45 g/L) in the water, which can form a common contaminant, MgSO_4 , which effect on aquatic toxicity has been studied by van Dam et al. (2010).

The wastewaters from the cyanidation process originate from detoxification processes. In the leaching simulation, the main concern of the wastewaters is the As concentration (0.31 g/L). This is the amount of arsenic that is not precipitated

in the pressure oxidation process. The As is predicted to be in As (III) form, which has high toxicity. Prior to releasing these waters into nature, the As (III) concentration needs to be lowered to acceptable safe levels of As(III), which in fresh water are 150 $\mu\text{g/L}$ (chronic) and 340 $\mu\text{g/L}$ (acute) (US EPA 2019). Similarly to wastewaters from pressure oxidation, also the waters from cyanidation have a high sulfate concentration (49.7 g/L) and alkaline pH around 8.9, precipitating, e.g., any dissolved Fe as hydroxides. In addition, the elution waters from gold recovery dilute the cyanide process water, and the impurities adsorbed by activated carbon are cleaned with hydrochloric acid. Suitable processing is needed for these waters prior to disposing them. The cyanide destruction (DETOX) is conducted after leaching, and success of this operation is critical in order to prevent CN^- ions release to the nature, severe consequences are evident for the aquatic life in case of cyanide leakage.

For halogen leaching process the same sulfate issue remains, as the sulfate concentration in brine solution is 58.27 g/L. However, in halogen process, the aim is to produce a brine that can be circulated back to the process. Thus, also the iron (0.71 g/L) and chloride (70 g/L) concentrations are high. In addition, the brine is acidic. If some of these waters are not circulated for chloride recycling, the process solutions are required to be subjected to the state-of-art metal precipitation (e.g., as sulfides) and neutralization.

4.3.6 Sensitivity analysis

The results of any LCA study are particularly dependent on the inventory data used and the assumptions made in the study (Norgate and Haque 2012). In this study, the LCA inventory was produced based on the best available data gathered from published sources for simulation of gold processes to produce a mass balance. The electricity consumption was calculated based on the HSC Sim mass balance and known values from the literature. The equations and constants used in the electricity calculations are presented in the ESM S1.4.

A sensitivity analysis was conducted on the benchmark scenario by varying the consumption of electricity and water by 10 to 20%. Variation of electricity consumption has a higher effect in halide leaching than in cyanidation in all impact categories. ± 10 to 20% variation in electricity causes ± 2.5 to $\pm 5.1\%$ effect on GWP in halide leaching and ± 2 to $\pm 4\%$ variation in cyanidation. The pressure oxidation contributes more on the overall variation value, due to the higher volume of the unit process compared to leaching and recovery stages. Similarly, the sensitivity to AP and EP varies from ± 4.8 to $\pm 7.1\%$ and ± 3.6 to 9.6% in halide leaching and $\pm 4.5\%$ to $\pm 9.1\%$ and ± 3.8 to $\pm 7.5\%$ in cyanidation.

By varying the water consumption in the processes, the sensitivity on estimated impacts is lower. In the cyanidation, ± 10 to 20% variation leads to ± 0.7 to $\pm 1.4\%$ change in GWP, ± 1.25 to

$\pm 2.5\%$ in AP, ± 2.6 to $\pm 5.1\%$ in EP, and ± 0.9 to $\pm 1.8\%$ in water depletion. For halide leaching, these variations are slightly larger; ± 0.74 – 1.5% (GWP), ± 1.1 – 3.95% (AP), ± 2 – 2.2% (EP), and ± 0.85 – 1.7% (water depletion). Similarly, in both processing routes, the pressure oxidation contributes more on sensitivities.

The effect of geographical location was investigated by comparing the effect the production region of electricity had on the process in high gold production countries, e.g., Australia and the USA. The electricity grid mix variation impacts all investigated environmental impact categories, Table 7. The global warming potential increases from the benchmark Finland scenario for ca. 35–40% if the electricity is produced in Australia or China. In addition, when compared to Finnish base values, the acidification potential and eutrophication potential increase significantly. Both the Australian and Chinese electricity grid mix will increase the estimated AP or EP over 100%. The electricity grid mix of USA has the largest impact on water depletion indicator with an increase of nearly 30%.

5 Discussion

The challenge in the simulation was the limited amount of available industrial information about halogen leaching, as it is only at the development stage. Although there are several publications available on the halogen leaching of gold, most of them describe leaching on laboratory scale. Thus, the parameters used in halogen simulation may not be the optimized parameters for an industrial-scale process. The mass balance simulated in this study may predict higher water consumption and chemical consumption compared to what can be achieved on larger scale since (i) water from condensed steam is not recovered and (ii) leaching residue washing is not optimized for the halogen process. An evaporation step is simulated for the halogen leaching, which is not officially reported for the process in literature. Evaporation can be energy consuming, which is observed from the mass balance (Table 6).

However, electricity consumption to produce the required heating energy for this unit was not included in LCA estimations as the definition of equipment used for the unit process was uncertain. In addition, the estimated LCA would not describe the results for the simple published process, which initially did not include evaporation unit process. Additionally, the steady-state simulation does not take into account the retention times and reaction times of concentrate in leaching. As a result, it is hard to evaluate the benefit of fast kinetics in the halogen process compared to cyanidation, which can require leaching times of over 24 h. This has an impact on the reactor sizes needed and thus the power consumption of the process, as significantly larger equipment is required. The reported leaching times for the halogen process can be as little as 1 to 2 h.

The study was also limited by the LCA database used. The NaCN chemical and Na₂SO₃ chemicals were cutoff flows in the assessment as upstream process was not found in the database. The addition of NaCN to SCE1 environmental indicator estimations can increase the impact values of AP and EP of cyanidation.

However, the simulations built in this study can be modified according to specific requirements of existing processes to produce more accurate data. Thus, the life cycle assessment can also be improved according to increasing knowledge about the development stage processes. This unbiased study shows that, based on the literature, simulations of development-stage processes can be built and environmental indicators can be predicted. This is essential, since the emerging technologies should be assessed as early as possible for their environmental impact. There are still unresolved problems related to cyanide-free gold leaching concerning gold recovery and water balance. However, if these problems can be overcome, a more competitive alternative can be attained. In addition, this study has focused on evaluating the simulation and environmental indicators obtained, and a more comprehensive sustainability assessment could highlight even more differences between the scenarios in the light of costs and social impact.

Table 7 The effect of electricity grid mix locality to benchmark (Finland) values as percentage increase

Country	GWP, CO ₂ -e		AP, H ⁺ mole e.		EP, N mole e.		Water depletion, kg	
	SCE1	SCE2	SCE1	SCE2	SCE1	SCE2	SCE1	SCE2
Finland (benchmark values)	9.3	11.1	10.9	16.1	26.6	46.6	60.6	76.7
Australia	35.3%	38.7%	150.8%	111.6%	170.6%	151.6%	3.1%	2.9%
USA	18.2%	21.6%	11.5%	8.6%	55.0%	50.9%	26.7%	29.2%
China	35.3%	40.5%	109.2%	84.8%	162.4%	150.9%	20.3%	22.2%

6 Conclusions

The current study presents refractory gold concentrate pressure oxidation and two different leaching process simulations: state-of-art cyanidation (SCE1) and cyanide-free halogen process (SCE2), still at the development stage. Cyanidation simulation predicted 98.5% gold recovery, whereas the halide leaching indicated a final gold recovery of only 87.3%, due to gold lost in the leach residue and wash waters along with the remaining cake moisture. However, with the addition of a secondary gold recovery unit for the leach residue wash water (SCE2b), gold recovery could be increased in halide leaching up to 98.5%. The halogen simulation predicted the consumption of 5 kg NaBr and 140 kg NaCl per ton of concentrate. The brine from evaporation could be used to lower the consumption of fresh chloride chemicals. The acidification and eutrophication potential have significant impact through NaCl production, which can be minimized with efficient recycling of the chloride chemicals.

The estimated environmental impacts predict that the development-stage cyanide-free process still has some challenges compared to cyanidation, as in the investigated scenarios the environmental impacts were generally higher for halogen leaching. This is mostly due to the lower recovery of gold (SCE2) in the process, as the environmental indicators are estimated based on the functional unit of 1 kg of Au. The simultaneous leaching and recovery on the activated carbon in carbon-in-leach process (SCE1) prevents gold losses to the leach residue and wash waters, since it uses a reagent (NaCN) that can be decomposed chemically (detoxification). The investigated halide leaching (SCE2) was based on separate leaching and recovery units, which results in gold losses to the moisture of the leach residue and wash waters. The washing of chlorides out of the leach residue further challenges the process water balance, leading to water accumulation. In the current study, the global warming potential of refractory gold concentrate treatment by POX and cyanidation was 10.3 t CO₂-e/kg Au, whereas POX followed by halogen leaching resulted in a 22% higher GWP, 12.6 t CO₂-e/kg Au. However, if the issues regarding gold recovery can be overcome in the halide process (SCE2b), a more competitive alternative will be obtained, lowering the GWP to 11.5 t CO₂-e/kg Au. Further process improvements, for example in the form of decreased moisture in the feed for halide leaching, and the adaptation of in situ gold recovery practices in chloride leaching as well, may give the cyanide-free processing options a competitive edge.

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