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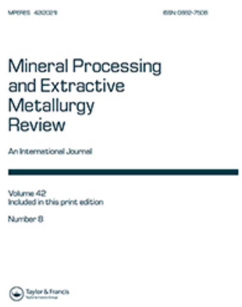

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Direct Cupric Chloride Leaching of Gold from Refractory Sulfide Ore: Process Simulation and Life Cycle Assessment

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ABSTRACT

A simulation-based life cycle assessment was conducted to compare the impacts of different cyanide-free chloride processing routes for refractory ores to determine whether one-stage leaching of gold from the ores could be advantageous over chloride leaching after conventional pressure oxidation from an environmental perspective. It was shown that both leaching conditions and the refractoriness of the ore strongly affect the environmental impacts of the process. The global warming potential in the direct leaching route was determined to vary between 7.7–17.0 t CO_2-eq for 1 kg gold/Au. Prerequisites for the sustainable application of the direct leaching process are suggested.

KEYWORDS

Sustainability; environmental impacts; non-cyanide lixiviants; halide leaching; process development

1. Introduction

High-grade gold ores that are highly amenable to conventional cyanide leaching have been largely depleted over time. This has a significant effect on the environmental footprint of gold production as refractory ores cannot be easily treated by simple cyanidation treatment. In refractory ores, gold is typically encapsulated inside a sulfide matrix (pyrite, arsenopyrite) that must be decomposed to liberate the gold for leaching (Marsden and House 2005). More complex treatment procedures and additional pre-treatments such as pressure leaching, bio-oxidation, or roasting, are typically utilized to attain sufficient gold recoveries (Lunt and Weeks 2005), which further increases the environmental burdens of gold processing from refractory resources (Norgate and Haque 2012).

The ability of acidic chloride solutions to dissolve gold and refractory sulfides simultaneously has been recognized as a potential advantage of gold leaching (Lundström et al. 2015; Miettinen et al. 2013), but this has been demonstrated in only a few experimental studies using very strong oxidants such as hypochlorite or bromate (Hasab, Raschi and Raygan 2013, 2014; Wang et al. 2019). In addition to the aforementioned hypochlorite and bromate, other oxidants such as hydrogen peroxide, ozone, and ferric salts, are known in the literature (Aylmore 2005). The use of cupric or ferric salts as oxidants has the advantage that the regeneration of the oxidant can be easily achieved by dissolved oxygen in the leaching reactor (McDonald et al. 1987; Senanayake 2004). Alternatively, chloride solutions can also be used for gold leaching after conventional pressure oxidation. Ahtiainen (2020) demonstrated the direct leaching of gold from refractory and preg-robbing double refractory ores in cupric chloride-bromide solutions, and the raw material characteristics were shown to affect the extraction and sulfide oxidation significantly. In addition to sulfidic gold encapsulation, native carbonaceous matter, which adsorbs dissolved gold, may cause gold losses due to the phenomenon called preg-robbing (Helm et al. 2009). In the future, chloride solutions may provide an alternative to toxic cyanide, which is dominantly used in industrial gold plants despite the extensive search for safer substitutes, such as thiosulfate, thiourea, and halides (Hilson and Monhemius 2006; Konyratbekova, Baikonurova and Akcil 2014; Melashvili et al. 2016).

The environmental impacts of gold production by conventional cyanidation have been studied in several life cycle assessments (LCA) for both refractory and free-milling ores (Cenia et al. 2018; Chen et al. 2018; Norgate and Haque 2012). Gold has a high specific environmental footprint, which arises mainly from the fact that gold concentrations in ores are very low, at ppm level. Therefore, massive amounts of energy is expended on mining and comminution, followed by pressure oxidation in the case of refractory ores (Norgate and Haque 2012). In the open literature, there are also some LCA studies on halide systems (Elomaa et al. 2020a, 2020b), and it appears that most of the environmental impacts of refractory ore treatment are caused by the pressure oxidation treatment. It has been suggested that direct leaching (avoiding pressure oxidation) of refractory gold ores could be technically feasible under atmospheric conditions (Hasab, Raschi and Raygan 2013, 2014; Lundström et al. 2015; Miettinen et al. 2013; Wang et al. 2019); however, the holistic evaluation of the process and specifically its environmental impact is lacking. This study investigates the environmental impacts of direct cupric gold chloride leaching from refractory ores, in order to provide numerical and comparable environmental impact values for the alternative development-stage processes for gold processing. The results are also compared with the other potential cyanide-free processing route: pressure oxidation followed by gold chloride leaching.

In this study, environmental impacts were investigated using simulation and LCA tools. The process simulation was conducted using Outotec's HSC Sim 9.0 (Outotec 2020) based on experimental data gathered from the literature. This allowed the

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calculation of the mass and energy balance of the process, which was adapted into a life cycle inventory (LCI). Life cycle assessment is a standardized framework for assessing the environmental impacts of a product or a process (ISO 14040 2006a, 2006b), and a similar process model-based approach has been validated in several earlier metallurgical studies (Abadías Llamas et al. 2019; Elomaa et al. 2020a, 2020b; Fernandes, Abadías Llamas and Reuter 2020; Pell et al. 2019; Reuter 1998). The life cycle impact assessment (LCIA) was conducted using GaBi software (sphera 2020). Direct leaching was compared against pressure oxidation followed by cupric chloride leaching by adjusting the earlier models of Elomaa et al. (2020a, 2020b).

2. Materials and methods

The study investigates the environmental impacts of gold processing via simulation-based methodology. The method consists of process simulation, which is carried out based on experimental work or literature data. The inputs and outputs from the process are compiled into a life cycle inventory and the impacts are calculated.

2.1. Process flowsheet

Simulation-based life cycle analysis was used to assess the environmental footprint of direct gold leaching from refractory ores in more aggressive cupric chloride solutions. The modeled direct cupric chloride leaching flowsheet (Figure 1) is based on the process developed by Outotec (Miettinen et al. 2013) and the model of Elomaa et al. (2020a) with some adaptations. Direct cupric chloride leaching was compared against pre-oxidation followed by milder cupric chloride leaching dedicated for gold dissolution (henceforth referred to as the *Reference process*), as described earlier by Elomaa et al. (2020a) The direct chloride leaching system is described in detail in the supplementary material.

The main steps in the direct chloride leaching process are as follows: atmospheric leaching, gold recovery by solvent extraction (SX), gold reduction, and neutralization steps. In the *Reference process*, chloride leaching and gold recovery are preceded by sulfide breakdown at elevated temperature and pressure (190°C, 100 psi oxygen overpressure (Marsden and House 2005)), countercurrent decantation and neutralization, leaving gold liberated for cupric chloride leaching. Milder conditions can be used for chloride leaching in the *Reference process* due to the prior oxidation of the pyrite-arsenopyrite matrix (Ahtiainen 2020). Gold dissolution from free-milling ores has been reported to occur in chloride solutions as mild as sea water ($[Cl^-]$ = 20 g/L, $[Cu^{2+}]$ = 3 g/L) (Ahtiainen and Lundström 2019), but gold dissolution is significantly faster under more aggressive conditions.

In the direct leaching process, the concentrate is mixed into a slurry in pre-acidulation, where the leaching chemicals are also added to adjust the slurry chemistry and to dissolve acid consuming minerals, such as carbonates. The slurry is leached under atmospheric pressure at 95°C with continuous oxygen feed. The pH is maintained below 2.5 by the addition of hydrochloric acid to keep the copper soluble (McDonald et al. 1987). The leaching residue is washed with water and then disposed of. The filtrate is circulated back to leaching while the washing waters are subjected to solvent extraction together with the PLS. Gold is extracted from the PLS in solvent extraction using a combination of an organic diester and a long-chain alcohol as extractant (Haapalainen 2018; Paatero and Haapalainen 2015). Gold is stripped from the organic phase into a mild solution of NaCl (10 g/L), and gold is reduced into metallic gold with sodium metabisulfite ($Na_2S_2O_5$, SMBS) (Paatero and Haapalainen 2015). The reduced gold is filtered from the depleted solution, which is neutralized in the subsequent effluent treatment step by adding limestone ($CaCO_3$) or slaked lime ($Ca(OH)_2$). Impurity metals in the solution are precipitated as hydroxides and a gypsum ($CaSO_4$

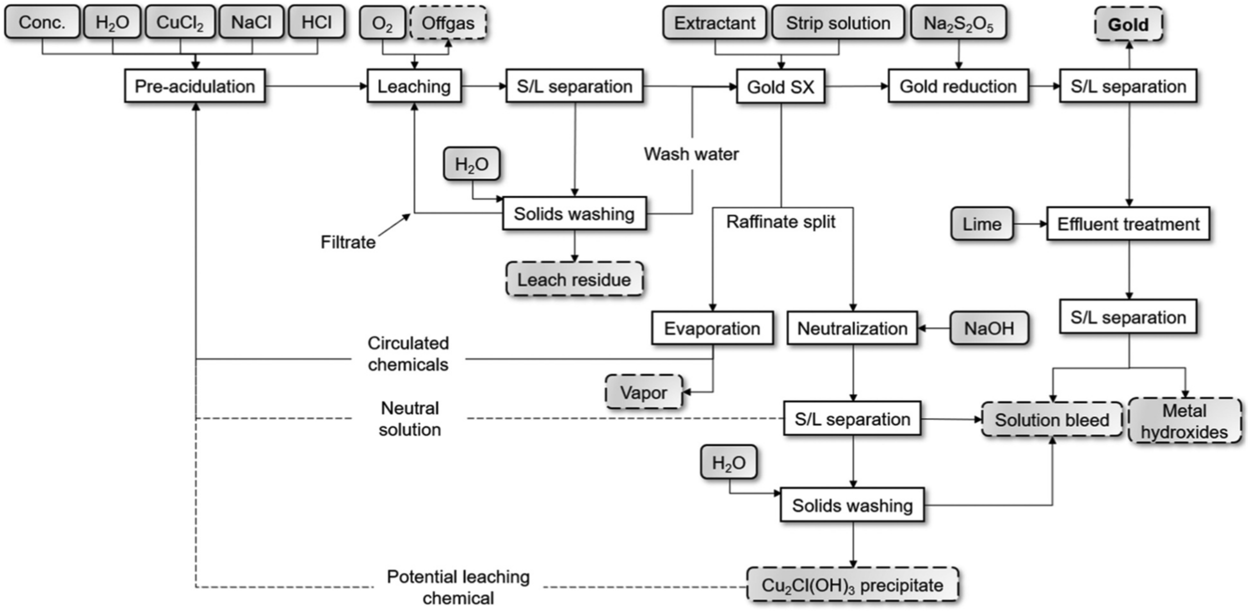


Figure 1. Direct cupric chloride leaching flowsheet for refractory gold concentrate.

$\cdot 2H_2O$) cake forms. The solid residues are disposed of and the treated solution is routed to further wastewater treatment. The raffinate from solvent extraction is divided between the neutralization and evaporation steps. In evaporation, the solution containing copper, acid, and chloride is concentrated and recycled back to pre-acidulation to decrease the consumption of chemicals. In neutralization, the solution is treated with NaOH to produce a precipitate that consists of atacamite ($Cu_2Cl(OH)_3$) and impurity hydroxides. The atacamite residue has the potential to be used as a source of cupric ions and chloride again in the leaching stage. The neutralized solution can be circulated back to the leaching step directly or, if necessary, through an additional sulfate removal step.

2.2. Process simulation

2.2.1. Raw material

The selected refractory ore minerology for the simulation is presented in Table 1. The gold and silver content in the concentrate was 38.7 and 33.0 ppm, respectively. In the raw material, 70 wt.% of gold was distributed in arsenopyrite (FeAsS) and 30 wt.% in pyrite (FeS_2) (Jaatinen 2011). The raw material could be assumed double refractory based on the original analyses due to the high native carbon content (1.95%) (Elomaa et al. 2020b), whereas the simulated process is not suited for preg-robbing raw materials. The native carbon in the analyses was replaced by inert quartz to avoid the preg-robbing issue. The throughput was determined by assuming that 10 tonnes of Au is produced annually with a minimum recovery rate of 80%. A typical year of 8000 hours of operation was assumed, resulting in a throughput of 40.2 t/h.

Table 1. Refractory gold concentrate composition, modified from Jaatinen (2011) by replacing C with SiO_2.

Mineral	FeS_2	FeAsS	$KAl_2(Si_3Al)O_{10}(OH)_2$	SiO_2	$CaCO_3$
wt.%	32.05	13.82	15.86	14.54	9.81
Mineral	$MgCO_3$	$NaAlSi_3O_8$	$FeTiO_3$	Fe_2O_3	$Cu_{10}Fe_2Sb_4S_{13}$
wt.%	5.34	3.25	2.85	1.67	0.34
Mineral	NiSb	Sb_2S_3	Au	Ag	
wt.%	0.22	0.19	38.7 ppm	33.0 ppm	

2.2.2. Process parameters

The controlled parameter values used as the basis for the models are provided in Table 2. Most of the parameters were controlled in the model, and where applicable, the controlling variables are also provided in the table.

2.2.3. Studied scenarios and cases

In the study, the mass and energy balances were compared between the two mineralogical scenarios to assess the effect of sulfur oxidation and leaching conditions. In the *Total oxidation* scenario (100% sulfide oxidation) gold was assumed to be very finely distributed in the sulfides, and gold leaching was assumed to be comparable to the dissolution degree of the host sulfides. Almost linear dependencies between sulfide oxidation and gold leaching efficiency have been observed with several ores (Ahtiainen 2020; Linge and Welham 1997; Pangnum and Browner 1996). However, the actual extent of sulfide oxidation required for gold liberation is individual to each ore (Marsden and House 2005), therefore, the *Partial oxidation* scenario (50% sulfide oxidation) was also investigated. High gold recoveries can be achieved by partial oxidation only in cases where gold forms clusters in the ore or is associated with porous sulfide particles, as in the case of the ore investigated by Bidari and Aghazadeh (2018). In such a *Partial sulfide oxidation* scenario, high gold extraction can be expected with incomplete sulfide oxidation, i.e., sulfide oxidation and gold dissolution are not linearly dependent on each other.

Gold leaching is also highly dependent on the aggressivity of the leaching conditions. Therefore, direct chloride leaching in two chloride leaching conditions with different chemical concentrations was investigated: the *Severe case* and the *Base case*. In the *Severe case* ($[Cu^{2+}]$ = 70 g/L, $[Cl^-]$ = 180 g/L, $[Br^-]$ = 5 g/L, T = 95°C, atmospheric pressure), aggressive leaching conditions are predicted to facilitate faster leaching of refractory sulfides (Elomaa et al. 2018). The expected disadvantage of the *Severe case* is the increased environmental burden due to increased chemical consumption (Elomaa et al. 2020a), although lower reactor energy consumption is expected due to the faster leaching kinetics (15 h in the *Total oxidation*

Table 2. Model parameters and the control variables in the simulation model.

Unit	Parameter	Control variable	Parameter value	Reference
Leaching	Cu^{2+} concentration	$CuSO_4$ input	>30 g/L	Elomaa et al. (2018)
Leaching	Cl^- concentration	NaCl input	>120 g/L	Elomaa et al. (2018)
Leaching	Slurry S/L	Split (%) to evaporation	300 g/L	Estimate
Leaching	Slurry S/L	TH2 overflow split (%)	75%	Estimate
Leaching	HCl concentration	$CaCO_3$ input	<20 g/L	Miettinen et al. (2013)
Leaching	Temperature	Cooling water input	95°C	Miettinen et al. (2013); Ahtiainen and Lundström (2019)
Leaching	pH	–	0.5–2.5	Miettinen et al. (2013); Ahtiainen and Lundström (2019)
Leaching	Oxidation potential	–	>450 mV v. SCE	Elomaa et al. (2018)
Leaching	Pressure	–	Atmospheric	Miettinen et al. (2013)
Gold SX	Au concentration in organic	–	3–10 g/L	Paatero and Haapalainen (2015)
Gold SX	Stripping NaCl concentration	H_2O input	10 g/L	Paatero and Haapalainen (2015)
Gold reduction	Sodium bisulfite: Au ratio	Sodium bisulfite input	2.25: 1 mol	Haapalainen (2018)
Gold reduction	Sodium bisulfite concentration	H_2O input	95 g/L	Haapalainen (2018)
Neutralization	pH	NaOH input	>2.5	Estimate
Effluent treatment	pH	$CaCO_3$ input	7	Estimate
Thickeners	Solids in thickener underflow	–	40%	Outotec (2020)
Filters	Washing efficiency	–	96%	Outotec (2020)
Filters	Washing water amount	H_2O input	1.5 m^3/t solids	Outotec (2020)
Filters	Cake moisture	–	25%	Outotec (2020)

scenario and 7.5 h in the *Partial sulfide oxidation* scenario). This could potentially mitigate the negative effects of high chemical consumption. In the *Base case*, less aggressive conditions are used (*[Cu^{2+}]* = 40 g/L, *[Cl^{-}]* = 125 g/L, *[Br^{-}]* = 5 g/L, *T* = 95°C, atmospheric pressure) and this is expected to facilitate the fast leaching of gold but also the sufficient leaching of refractory sulfides (Elomaa et al. 2018; Seisko, Aromaa and Lundström 2019). However, the leaching times are slightly higher (20 h in the *Total oxidation* scenario and 10 h in the *Partial sulfide oxidation* scenario), indicating an increase in energy consumption. It should be acknowledged that the retention times are estimated from a model obtained by electrochemical measurements (Elomaa et al. 2018), whereas in real processing even longer retention times may be necessary.

Comparison of the direct chloride leaching processes described above was conducted vs. the *Reference process*, consisting of pressure oxidation followed by cupric chloride leaching, a model from Elomaa et al. (2020a, 2020b). The more aggressive conditions described above were considered redundant for pressure oxidized gold concentrates. The *Reference process* in the current study consisted of autoclave leaching and the liberated gold was chloride leached in the *Base* conditions (*[Cu^{2+}]* = 40 g/L, *[Cl^{-}]* = 125 g/L, *[Br^{-}]* = 5 g/L, *T* = 95°C, atmospheric pressure) for comparison. The *Base* conditions were also evaluated in the study by Elomaa et al. (2020a).

2.2.4. Simulation assumptions

The simulation was conducted based without specific knowledge on how the selected raw material would behave in the described conditions. The experimental results used to predict the kinetics of sulfide dissolution were collected from simplified systems, and the complexity of the described system increases uncertainty in the model. It was presumed that the presence of ferric ions would likely further promote the dissolution of gold and other components in the raw material, but this was not included in the model.

Ahtiainen (2020) obtained high gold extraction levels (98%) from free-milling gold ores after five hours of leaching under milder conditions (150 g/L chloride, 15 g/L aqueous copper, 6.2 g/L bromide), but the findings for the direct leaching of refractory ores were less conclusive due to the effects of raw material behavior. The flowsheet presented in Figure 1 is not applicable for double refractory ores based on the study by Ahtiainen (2020) due to the occurrence of preg-robbing in chloride solutions. It is suggested that direct leaching could be a viable processing technology for some ores, especially if the gold extraction is not linearly dependent on the oxidation degree of the sulfides. It was shown by Ahtiainen (2020) that the dissolution of gold occurs far faster in chloride solutions than the oxidation of sulfides; thus it was presumed in the simulation that gold would dissolve completely when either 99% or 50% of the sulfide matrix were oxidized, depending on the scenario.

Although the effect of the Fe(III)/Fe(II) redox pair was not included in the simulation, it could have a significant effect on the consumption of copper in the system. Fe(III) is a stronger oxidant than Cu(II) and the dissolution of iron-bearing minerals from refractory ores introduces a large amount of iron into the solution. The iron contained in the raw material is used, for instance, in the direct atmospheric leaching of sphalerite concentrates (Lorenzo-Tallafigo et al. 2018).

Gold solvent extraction and reduction step extraction were based on published experimental literature (Paatero and Haapalainen 2015): 99.9% of gold was assumed to be recovered into the enriched electrolyte, and 99.9% of gold in the electrolyte was recovered as metallic gold in the subsequent reduction step. Solvent extraction was simulated as a black-box distribution model without including the organic phase at all in the simulation. This was done due to the lack of applicable organic extractants in the HSC database. Solvent extraction is also sensitive to solids, so an additional clarifying settler would likely be needed in industrial practice. Thickeners and filters were modeled with HSC Sim's inbuilt constants.

It must be emphasized that high gold extraction in leaching (99.7%) was assumed in all the simulated cases and scenarios, which influences the results of the LCA. The extraction percentage was selected to be the same as that presented in the model by Elomaa et al. (2020a) to obtain comparable values. Overall, the results are indicative of the best-case scenario, and lower gold extraction or slower host sulfide dissolution would inevitably lead to higher environmental impacts of the process. Further kinetic data is consequently needed to evaluate the impacts of direct refractory gold ore leaching more accurately.

2.3. Scope and boundaries in the life cycle assessment

The functional unit in the study was 1 kg of gold, which was produced from a purified solution by reduction. The scope of the study is regional (Finland) and limited to the operational stage of the plant, which excludes steps such as the construction of the plant and equipment maintenance. The technical system boundary is gate-to-gate and presented in Figure 2. A comparison of both direct chloride leaching processes as well as the *Reference process* is included in the boundary.

The LCIs were generated using HSC Sim, which was used to calculate the mass and energy balances of the investigated gold processing options. In addition, electrical power consumption was calculated for all the key equipment (for different scenarios and cases) in the process based on the mass balance.

Sodium bromide and the organic phase in solvent extraction had no relevant upstream data and were left as cutoff flows. The Finnish average supply mix was used to represent electricity production. The cupric chloride used as the source of Cu^{2+} in the model of Elomaa et al. (2020a) was changed to copper sulfate in our models ($CuSO_4$), due to the lack of background data for cupric chloride in the simulations.

Global warming potential (GWP), acidification potential (AP), freshwater eutrophication potential (EP), and water depletion (Water) were assessed in the evaluation for both direct gold chloride leaching as well as for the *Reference process*. GWP describes the heat trapped in the atmosphere by the emitted greenhouse gases (CO_2, CO, CH_4, N_2O, etc.) with respect to CO_2. AP indicates the emissions that form acid rain in the atmosphere (mainly SO_2, NO_x, NH_3), while EP quantifies the amount of nutrients (P, phosphates) released to aqueous ecosystems. Water depletion estimates water consumption.

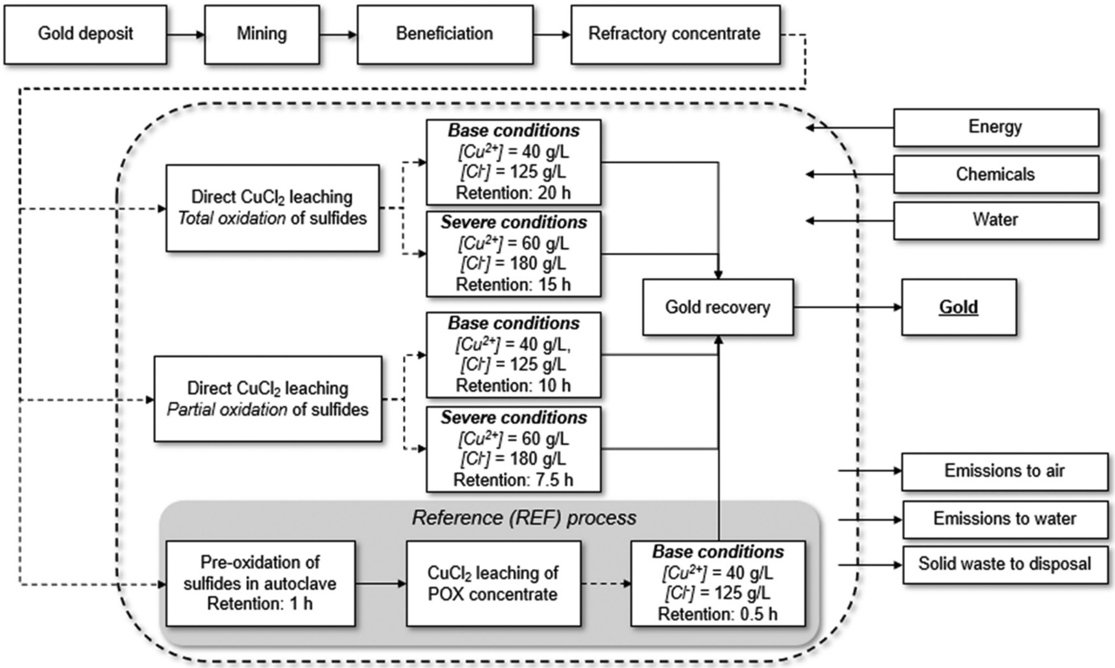


Figure 2. Simplified process flowsheet for gold production from ore by direct cupric chloride leaching in the compared scenarios and cases. The Reference process consisting of pressure oxidation and gold leaching from pressure oxidized concentrate is highlighted with a gray background. The technical boundary of the modeled system is represented by the dashed line.

3. Results and discussion

The feasibility and potential problem areas of the direct leaching process were evaluated in both the simulation and the LCA. Simulation was useful in determining chemical consumption, waste burdens, water balance, and the accumulation of compounds in the system, while the environmental impacts were quantified in the LCA.

3.1. Simulation results

The simulated flowsheet and mass balance of the process is presented in Figure 3. Due to the circulation of the solution in the investigated direct leaching models, aqueous sulfate was observed to quickly accumulate in the system, which is why the bleed streams from both gold reduction and SX raffinate treatment were gathered into an additional gypsum precipitation

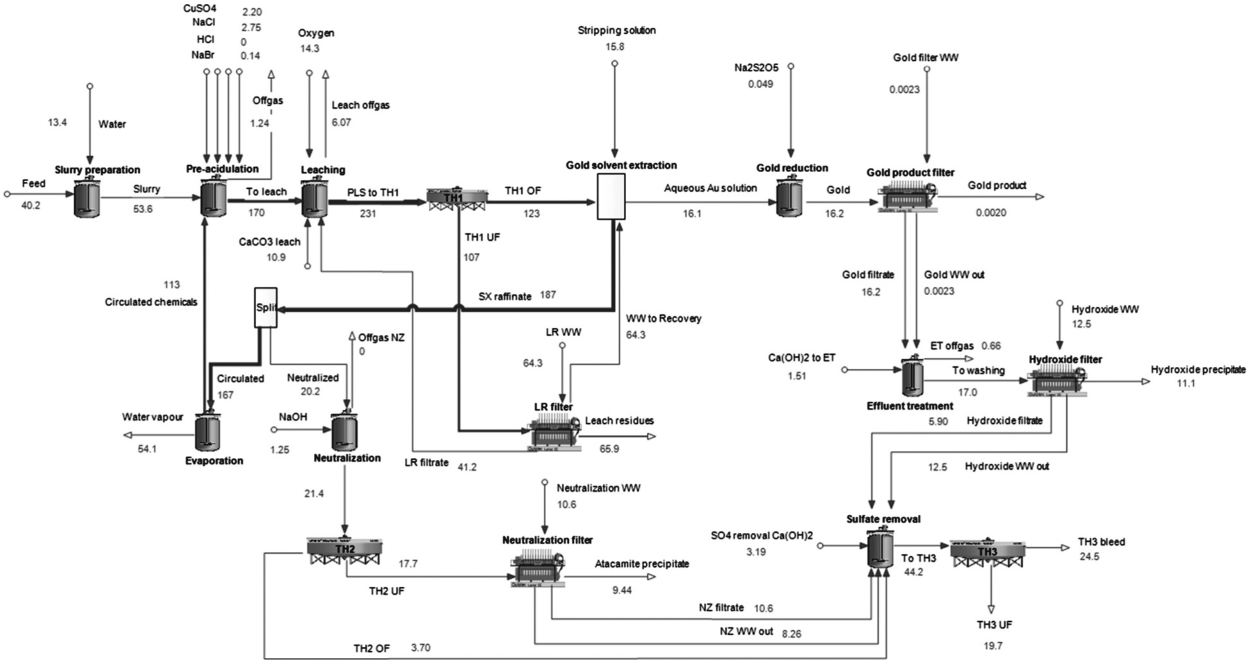


Figure 3. Mass balance of direct chloride leaching: total oxidation scenario Base case, with all streams expressed in t/h.

unit. The sulfate levels in the combined wastewaters could be lowered to approximately 10 g/L this way. The oxidation of sulfides also generated excessive amounts of acid, which is why the acid levels in leaching were controlled to the maximum level of 20 g/L as HCl, corresponding to pH 0.1, using limestone to neutralize the solution.

3.1.1. Chemical consumption

In direct chloride leaching, both the extent of sulfide oxidation and the leaching conditions were shown to have a significant effect on the consumption of leaching chemicals, which can be observed in Table 3. As expected, *Severe* conditions consumed chemicals more than *Base* conditions, but the difference between *Total* and *Partial oxidation* was also distinct. In *Partial oxidation*, the consumption of neutralizing chemicals was only a third of the consumption in the *Total oxidation* scenario, which is explained by the lower extent of sulfide oxidation to sulfuric acid. In *Severe* conditions, the effluents contain more metals such as Cu and Fe, which is why slightly more neutralizing chemicals are consumed in the effluent treatments than in *Base* conditions.

Controlling the pH of the PLS in leaching by adding limestone led to a high consumption of neutralizing chemicals in the *Total oxidation* scenario. When pressure oxidation was also considered, the overall consumption of $CaCO_3$ and NaOH was 430 kg/t feed (Elomaa et al. 2020a, 2020b). In the *Partial oxidation* scenario, almost all of the raffinate (95–96%) is circulated via evaporation, which considerably decreases consumption of NaOH and NaCl especially. The consumption of leaching and neutralizing chemicals could also be decreased in both scenarios by using the $Cu_2Cl(OH)_3$ rich neutralization residue as a leaching chemical.

3.1.2. Water balance and process residues

In the model, the raffinate from solvent extraction was divided between the neutralization and evaporation steps, and the leaching chemicals were circulated back to leaching through evaporation. Miettinen et al. (2013) also circulated the copper-depleted, neutralized effluent back to leaching through evaporation. However, this was observed to lead to excessive consumption of cupric chloride in the current direct leaching model and therefore the neutralized effluent was bled out of the system instead. Elomaa et al. (2020a) controlled the slurry solids content earlier by the split ratio, and this approach was observed to lead to the minimum leaching chemical consumption in the current model as well. In the initial slurry preparation, the slurry solids content was set at 75% to introduce freshwater into the process.

Table 3. Direct chloride leaching chemical consumption, chemical circulation, and gold recovery in the modeled scenarios and cases.

	Total oxidation		Partial oxidation	
	Base	Severe	Base	Severe
Chemical consumption (kg/t feed)				
NaCl	61.9	95.4	29.1	51.7
$CuSO_4$	49.0	90.6	34.8	66.9
NaBr	3.1	3.3	2.3	2.4
NaOH	21.4	31.6	10.1	16.9
$CaCO_3$	310.7	329.1	100.8	115.2
Recovery of chemicals				
Split to evaporation (%/%)	90/10	89/11	96/4	95/5
Evaporated water (t/h)	68.1	67.6	64.0	63.0
Gold recovery (%)	99.2	99.2	99.2	99.2

The leaching residue consisted of inert oxides from the raw material, precipitation products of iron ($FeAsO_4$, Fe_2O_3, $NaFe_3(SO_4)_2(OH)_6$), and undissolved pyrite and arsenopyrite in the case of *Partial oxidation* scenario. The residues also contained 350 ppm Cu and 80 ppm As in the remaining moisture, which could make them hazardous waste depending on regulation. Large amounts of gypsum sludges (0.8 t/t_{feed} in *Total oxidation* scenario) were also formed due to the use of limestone and lime as the main neutralizing chemicals. Due to the metals and chloride mixed with the gypsum (25,000–35,000 ppm), the sludges cannot realistically be valorized in practical applications, such as in construction, and the gypsum would also have to be stored in tailing ponds (Nurmesniemi 2018). The high chloride content in the sludges is a problem that should be addressed by further filtering and washing of the waste, which would also result in more processing costs and aqueous process solutions. The chloride-containing filtrate and possibly wash waters could, however, likely also be re-used in the process, which would further improve the water balance. The moisture retained by the various residues produced in the process could also be pumped from the tailing ponds to decrease freshwater usage.

Acid mine drainage and potential arsenic release from the solid residues is a concern, particularly in the *Partial oxidation* scenario, due to the large amount of arsenopyrite and pyrite in the residues. Since arsenic mobilization from arsenopyrite is favored by low pH, acid mine drainage should be managed by neutralization (de Andrade et al. 2008). Another promising method for controlling sulfide oxidation would be stabilization with a mixture of cement and gypsum sludge, although the method has not been validated in long-term studies (Demers et al. 2015). The sulfides could be separated from the rest of the residue by flotation, which would reduce the volume of stabilized material.

3.2. Life cycle assessment results

The LCI for the studied processes and conditions were compiled from the simulation and used to calculate the impact categories. The sensitivity of the results to various process parameters and gold recovery was analyzed to assess the reliability of the model and to study the critical data gaps.

3.2.1. Inventory analysis

The LCIs for each of the direct leaching cases and scenarios are presented in Table 4. Updated LCI data for the *Reference process* in *Base* conditions is also presented in Table 4, and the data for pressure oxidation was directly taken from Elomaa et al. (2020a). The normalized data for 1 kg of gold was transferred to GaBi software, and data from the ecoinvent and GaBi professional databases were used in defining the background processes.

3.2.2. Environmental impacts

The environmental impact indicator values and the contributions of the background processes are presented in Figure 4a–d. With the adjusted LCI and the inclusion of copper sulfate, the GWP for the *Reference process* was 15.1 t CO_2-eq/kg_{Au}, whereas

Table 4. Life cycle inventory for the investigated direct cupric chloride leaching scenarios and cases as well as the *Reference process*, normalized for 1 kg of gold.

	Total oxidation		Partial oxidation		REF	
INPUTS	Base	Severe	Base	Severe	Base	Unit
Concentrate	27570.1	27570.1	27570.1	27570.1	21024.7	kg
Electricity	3853.8	3206.3	2808.4	2698.9	1152.5	kWh
$CuSO_4$	1348.5	2510.0	954.6	1849.1	1274.2	kg
HCl	0.0	0.0	0.0	0.0	192.5	kg
NaCl	1695.6	2650.2	787.1	1321.8	1433.5	kg
NaOH	587.4	941.3	267.0	467.3	398.2	kg
SMBS	0.6	0.6	0.6	0.6	0.6	kg
Lime	8578.1	9072.1	2717.0	3177.6	1486.6	kg
Oxygen	13810.1	13810.1	5694.3	5694.3	8.6	kg
Process water	144592.8	148264.4	95867.8	99899.9	68419.1	kg
OUTPUTS						
Gold	1.0	1.0	1.0	1.0	1.0	kg
CO_2	5026.7	5688.3	2696.9	2810.4	657.0	kg
Solid waste	48543.1	50550.7	36246.7	37935.6	25096.2	kg
Wastewater	141854.1	146311.7	94961.6	99240.7	83737.0	kg
Water vapor	307.1	440.6	201.4	251.2	0.0	kg

the originally calculated value was 12.6 t CO_2-eq/kg_{Au} (Elomaa et al. 2020a). The difference is explained principally by the addition of copper to the LCI data. In the direct chloride leaching *Total oxidation* scenario, the GWP values for the *Base* and *Severe* cases were 12.9 and 17.0 t CO_2-eq/kg_{Au}, respectively. In the *Partial oxidation* scenario, the corresponding values were 7.7 and 10.4 t CO_2-eq/kg_{Au}.

It can be clearly seen that out of the studied processes and conditions, direct chloride leaching – *Partial oxidation* in milder (*Base*) leaching conditions is the most beneficial environmentally and results in the lowest GWP, AP, EP, and water depletion values. Additionally, *Severe* direct leaching conditions also appear to provide lower environmental impacts compared to the *Reference process* in *Partial oxidation*. On the other hand, the direct leaching process consisting of *Total* sulfide *oxidation* in *Severe* conditions has the overall highest impact compared to the other investigated systems, including the *Reference process*. *Total oxidation* in *Base* conditions performs better than in the *Reference process* in terms of GWP but results in higher impacts in all other studied impact categories.

The results highlight the fact that both chloride leaching conditions and the state of sulfide matrix oxidation have a significant effect on the sustainability of the gold process. Selecting an environmentally optimal process for refractory gold ores is consequently highly dependent on the raw material characteristics. The direct leaching process appears more suitable for moderately refractory sulfide ores.

The direct CO_2 emissions from the use of limestone as the primary neutralizing chemical is the hotspot in the process in terms of GWP (27–45% of total GHG). Copper sulfate consumption also accounts for a large share (22–46% depending on the scenario) of the GWP, and alone explains well over 90% of the impacts in the AP and EP impact categories in all scenarios and cases, including the *Reference process*. In terms of water depletion, the impacts come mainly from copper sulfate and electricity production, while the other utilities and chemicals contribute very little to the overall environmental profile of the process. Based on the contribution analysis, the minimization of copper consumption by the optimization of effluent circulation and the use of the atacamite precipitate as a leaching chemical can provide the greatest benefits in the cupric chloride leaching process of gold. This issue could be avoided for high-copper gold ores, where

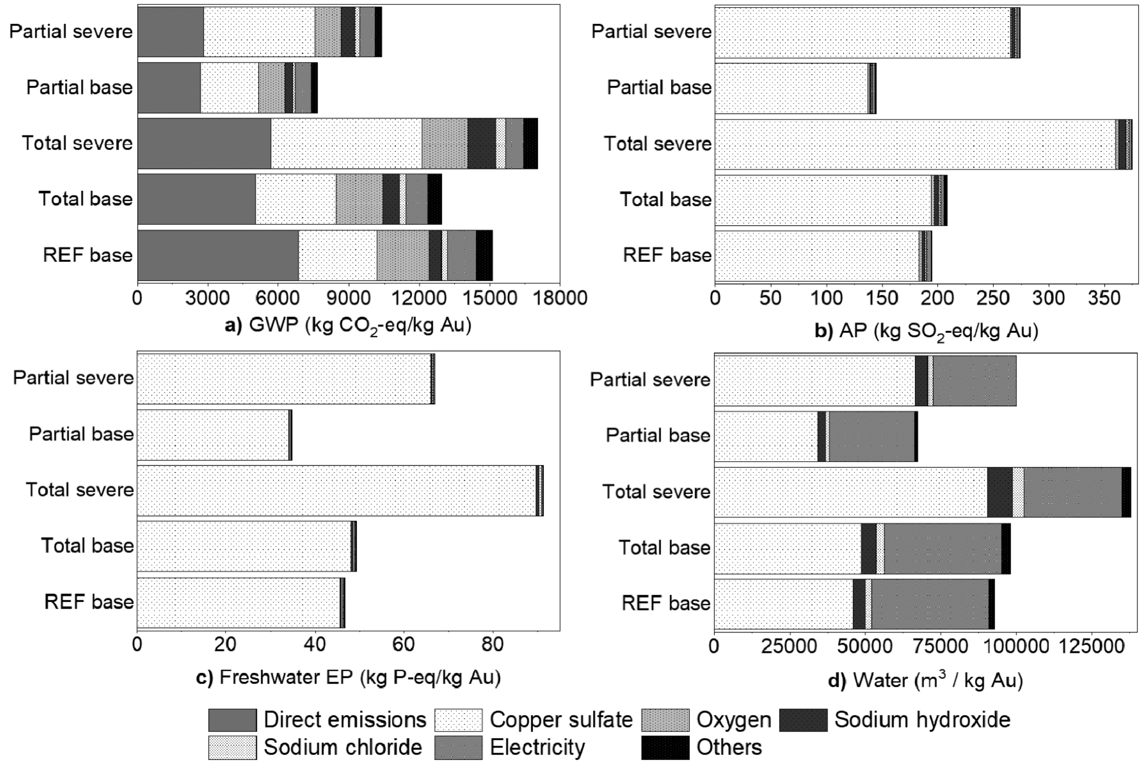


Figure 4. The environmental impacts of the direct cupric chloride leaching of refractory gold ore as well as the Reference process (pressure oxidation followed by leaching in Base conditions). (a) Global warming potential (GWP). (b) Acidification potential (AP). (c) Freshwater eutrophication potential (EP). (d) Water depletion (water).

the dissolution of the raw material can provide cupric ions for leaching and thus minimize copper consumption in the process (Dreisinger 2005).

The effect of electricity consumption on the environmental footprint of the process is smaller than typically expected from a metallurgical process, considering that shifting to clean energy is considered one of the key ways to decrease the environmental burden from the metallurgical and particularly the gold industry (Chen et al. 2018; Farjana et al. 2019). There are two reasons for this: first, the highly electricity-consuming mining and beneficiation stages were omitted in this study, and second, a comparably low-carbon average Finnish electricity supply mix was used as the electricity source.

The state-of-the-art process, which consists of pressure oxidation and cyanidation, has been studied by Elomaa et al. (2020b) for the same raw material composition, yielding a GWP value of 10.1 t CO_2-eq/kg_{Au}. Although further optimization of the chloride-based alternatives is possible, cyanidation appears difficult to compete against from a life cycle perspective. Cyanidation is operated at low cyanide concentrations (typically 0.15–0.3 g/L for free-milling ores), using air for oxidative power (Marsden and House 2005). The rapid kinetics in cupric chloride leaching can provide the chloride-based alternative with a competitive edge over cyanidation in terms of reactor electricity consumption, but the effect of chemical consumption may be difficult to overcome. Furthermore, the results suggest that the difference between cyanidation and chloride leaching could be even larger in terms of acidification, eutrophication and water depletion based on the relative contribution of copper sulfate production. If the chemical consumptions can be optimized and the problems properly addressed, direct cupric chloride leaching (7.7–10.4 kg CO_2-eq) appears to be a possibly competitive treatment option for moderately refractory ores in comparison to pressure oxidation and cyanidation (10.1 t CO_2-eq) (Elomaa et al., 2020b). Although direct cyanidation of refractory ores is not possible, other less intensive pre-treatments such as bio-oxidation could also be feasible before either cyanidation or cyanide-free leaching, which brings the environmental competitiveness of direct leaching by cupric chloride further into question.

The largest obstacle for the adoption of alternatives to cyanide in gold leaching is not the environmental aspects but the cost competitiveness and the ease downstream processing. The high consumption of leaching chemicals, such as halides, thiosulfate or thiourea, usually lead to non-cyanide lixiviants being unfeasible in comparison to cyanide. For instance, the consumption of thiourea is over three times that of cyanide (Li and Miller 2006). In the current study, the consumption of copper sulfate and sodium chloride were estimated to be approximately 60 kg/t in the best case (*Partial oxidation, Base case*), which is several times the consumption of cyanide even for the most cyanide-consuming ores (Li and Miller 2006). Chloride and thiosulfate may find their uses in the treatment of refractory ores, however, such as complex copper ores (Dreisinger 2005; Zhang and Senanayake 2016), or carbonaceous preg-robbing ores in the case of thiosulfate (Zhang and Senanayake 2016).

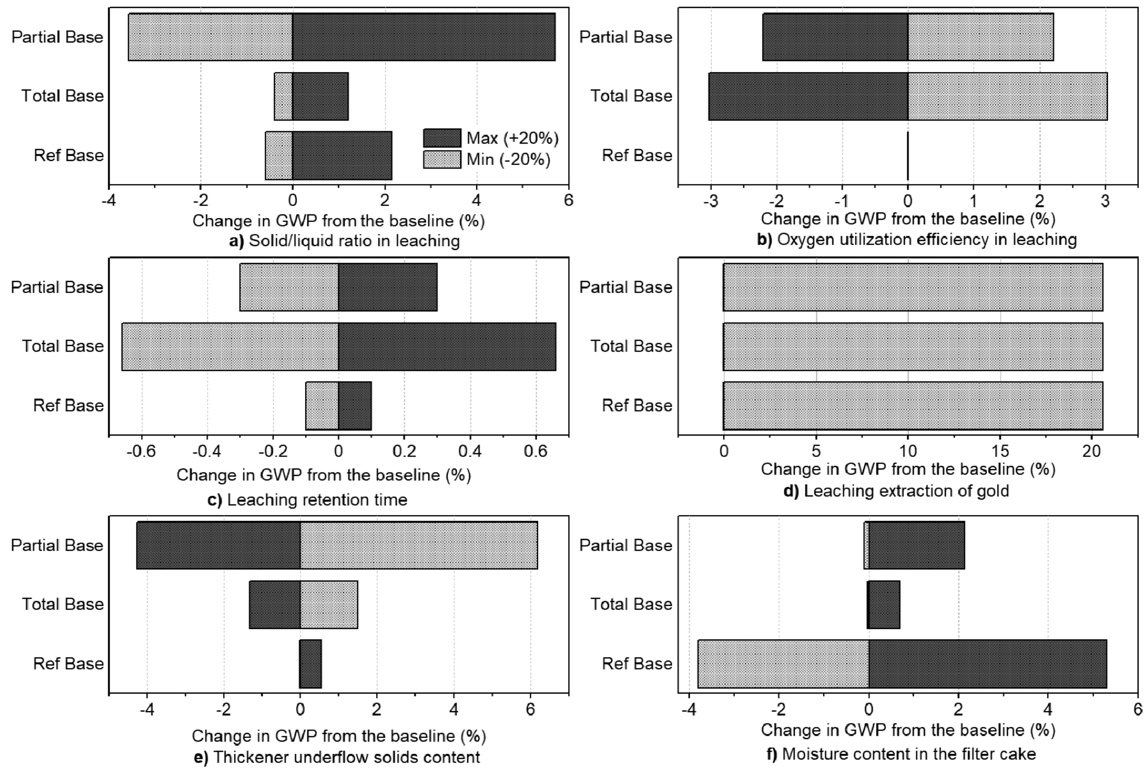
3.2.3. Sensitivity analysis

The sensitivity of several uncertain leaching parameters, such as Au extraction, the efficiency of oxygen utilization, retention time, and solid/liquid ratio, the solids content in thickener underflow (UF) slurry, and moisture content in filter cake, was assessed by changing the parameters in the simulation by 20% and measuring the change in the environmental indicators. The results of the sensitivity analysis for GWP are presented in Figure 5a–f and AP in Figure 6a–f. Values for also EP and Water are provided in the Supplementary material (Table S4). The direct chloride leaching *Base* conditions for each of the scenarios (*REF, Partial oxidation, Total oxidation)* were used to study the sensitivity of the system: 99.7% Au extraction, 80% oxygen utilization efficiency, 20 hours of retention time, 300 g/L solid/liquid (S/L) ratio, 40% solid content in thickener underflow, and 25% moisture in the filter cake. The evaluation was only done for the cupric chloride leaching model for the *Reference* due to the fact that pressure oxidation is an established and efficient pre-treatment for refractory ores. The sensitivity analysis was conducted to interpret the effect of the limitations in the model and estimate the importance of certain simulation parameters on the environmental profile of direct leaching in comparison to the *Reference process*.

Sensitivity analysis shows that the extent of Au extraction in leaching was strongly reflected in the normalized values for all of the inputs and outputs: a 20% decrease in gold extraction (~80% gold leaching) increased all of the environmental indicator values by more than 20%. This highlights that raw material characteristics are crucial in estimating the environmental performance of the process, and consequently even a minor decrease in gold extraction (compared to the *Reference* gold process) can easily make the benefits insignificant. Although this is also true for the *Reference process*, as seen in Figures 5 and 6, the recovery rates in the direct leaching process are far more uncertain than in the *Reference.*

Oxygen utilization efficiency in leaching was also included in the sensitivity analysis since excess oxygen is necessary due to the non-ideal mixing of the gas and consumption in the side reactions. The oxygen consumption was far higher in direct leaching when compared with the *Reference process*. The efficiency was estimated to be 80% in the simulation, but this is likely to be unrealistically high in atmospheric conditions even with highly efficient agitators. In pressure oxidation, oxygen utilization efficiency is typically 50–90% depending on the impeller design (Marsden and House 2005). However, the analysis shows that oxygen utilization only had an effect on the consumption of oxygen. Decreasing or increasing oxygen utilization efficiency affects GWP (±3.0% *Total oxidation*, ±3.0% *Partial oxidation*), but not the other categories to any significant degree. A far less significant effect was seen in the *Reference process* (±0.002%) due to the exclusion of the pressure oxidation step from the analysis, but efficient oxygen utilization is also expected from an autoclave. In the cupric chloride leaching step, oxygen was only consumed in the regeneration of the oxidant and some side reactions.

Retention time was one of the more uncertain parameters in the model due to the limited amount of data for industrial raw materials. The experimental work of Ahtiainen et al. (2020) on the direct leaching of gold would suggest that very long (up to



a) Solid/liquid ratio in leaching

b) Oxygen utilization efficiency in leaching

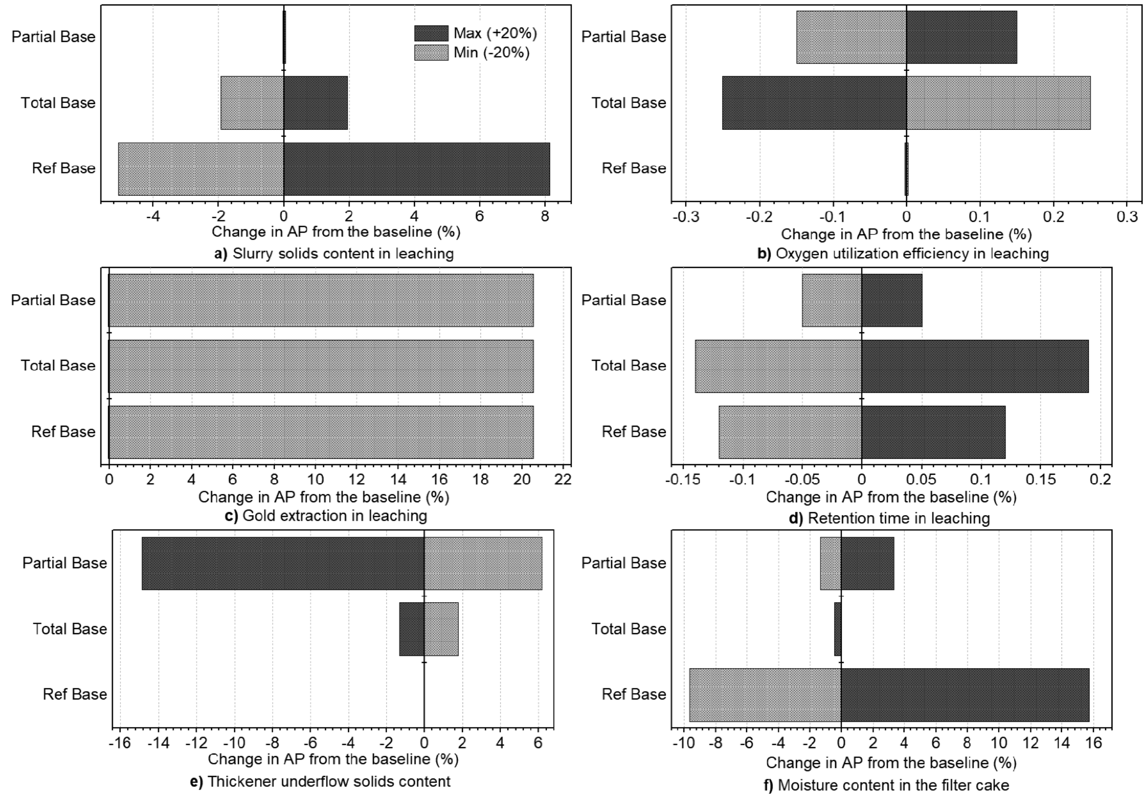
c) Leaching retention time

d) Leaching extraction of gold

e) Thickener underflow solids content

f) Moisture content in the filter cake

Figure 5. The sensitivity of GWP environmental indicator to 20% change in various process parameters. (a) Slurry solid/liquid ratio in leaching. (b) Oxygen utilization efficiency in leaching. (c) Gold extraction in leaching. (d) Retention time in leaching. (e) Solids content in the thickener underflow. (f) Moisture content in filter cakes.



a) Slurry solids content in leaching

b) Oxygen utilization efficiency in leaching

c) Gold extraction in leaching

d) Retention time in leaching

e) Thickener underflow solids content

f) Moisture content in the filter cake

Figure 6. The sensitivity of AP environmental indicator to 20% change in various process parameters. (a) Slurry solid/liquid ratio in leaching. (b) Oxygen utilization efficiency in leaching. (c) Gold extraction in leaching. (d) Retention time in leaching. (e) Solids content in the thickener underflow. (f) Moisture content in filter cakes.

60 hours) retention in leaching could be expected even under highly aggressive conditions (100 g/L copper, 225 g/L chloride, 78 g/L bromide). The increase of retention time by 20% in the *Total oxidation* scenario, *Base* case conditions is reflected in the electricity consumption, but only a modest effect was seen in all of the scenarios when the relatively clean Finnish electricity

supply mix is used: ±0.47% *REF* process, ±0.66% *Total oxidation*, ±0.30% *Partial oxidation*. This suggests that the consumption of leaching chemicals has a more profound effect on the environmental footprint of the process than electricity consumption in leaching. However, the effect of electricity consumption is expected to be more significant in geographical areas with more carbon-intensive energy supply mixes. Long retention times also increase the capital costs in setting up the plant, thus affecting the economic feasibility, although this was beyond the scope of the current study.

Increasing slurry solids loading was shown to increase the values for the environmental impacts, despite the fact that smaller amounts of leaching chemicals are typically needed with high S/L ratios. This occurred because the S/L ratio was controlled at the raffinate split, where the copper- and chloride-bearing aqueous solution was either recycled back to leaching or routed out of the process via neutralization. Increasing the solids loading in the slurry consequently resulted in less of the solution being recycled and a higher consumption of commercial grade chemicals. A particularly strong effect was seen in the *Partial oxidation* scenario where increasing S/L to 360 g/L doubled NaCl and NaOH consumptions due to the lower chemical circulation (+5.71% GWP, +4.70% AP). In the case of the *Reference process*, the S/L ratio also affected HCl consumption in the process in a similar manner, whereas excessive acidity of the leach slurry was an issue for direct leaching. The results suggest that the circulation of leaching chemicals is critical in developing an environmentally viable halide leaching process for gold.

The effect of thickener underflow slurry density and filter cake moisture on the process were less clear. It was observed from Figures 5 and 6 e that lower UF slurry density significantly increased the environmental impacts particularly in *Partial oxidation* scenario, whereas this had very little effect on the *Reference process*. This was possibly due to the accumulation of impurities that circulated in the process with the filtrate, as a similar effect was not seen in the *Reference process*. Moisture content in the filter cake was largely inconsequential for the direct leaching process, but the effect on the *Reference process* was significant (roughly ±5% GWP, ±>10% AP). The pressure oxidized gold concentrate slurry was washed in a CCD circuit and filtered prior to being subjected to cupric chloride leaching (Elomaa et al. 2020a), meaning that high moisture content in the filtered material diluted the slurry and less of the gold-depleted raffinate was circulated back to leaching and more chemicals were thus consumed.

4. Conclusions

The direct leaching of refractory gold ores in chloride solutions is a conceptual, unproven process, but it has been hypothesized to be able to provide a potential alternative to the costly and intensive pre-oxidation processes in the treatment of refractory gold ores. In this study, the environmental impacts of this development-stage processing option were evaluated at an early stage. It was calculated that the global warming potential for direct chloride leaching – *Base* leaching conditions in the *Total oxidation* scenario – would be 12.9 t CO_2-eq for 1 kg of gold, whereas *Severe* leaching conditions in direct chloride leaching would result in 17.0 t CO_2-eq with complete sulfide oxidation. In the *Partial oxidation* scenario, the corresponding values for *Base* and *Severe* conditions were 7.7 and 10.4 t CO_2-eq respectively, whereas the *Reference process* (pressure oxidation and cupric chloride leaching) yielded 15.1 t CO_2-eq/kg$_{Au}$. The results achieved indicate that the simultaneous oxidation of refractory sulfides and the leaching of gold in acidic cupric chloride solutions, i.e., direct chloride leaching could be an environmentally competitive alternative to the *Reference process*: pressure oxidation followed by gold leaching. However, this is only if certain prerequisites (i–iv) are met:

- The consumption of fresh leaching chemicals, particularly copper, should be minimized by the efficient recycling of the solvent extraction raffinate and the utilization of the copper-bearing atacamite precipitate.
- Clear benefits can be observed for moderately refractory gold ores. Direct chloride leaching is environmentally competitive for raw materials where only partial oxidation of sulfides is required to liberate gold.
- Complete oxidation of the refractory sulfides in cupric chloride solutions is only competitive under sufficiently mild leaching conditions (40 g/L copper, 125 g/L chloride, 5 g/L bromide); however, based on earlier studies, the efficient recovery of gold under such conditions appears unlikely.
- Based on the sensitivity analysis, gold extraction affects the impact categories proportionally and achieving high gold recovery is crucial to make direct chloride leaching competitive economically and environmentally. The process is consequently not suitable for preg-robbing ores without significant modifications.

The findings of this study suggest that the application of pressure oxidation for refractory gold ores is environmentally more attractive for highly refractory ores due to the presumably high consumption of leaching chemicals, but there could be clear benefits in direct leaching for partially refractory sulfide ores depending on the leaching conditions. The sensitivity analysis suggests that efficient oxygen dispersion in the direct leaching reactor has a significant effect on the GWP values from the process, whereas the effect of electricity consumption and the solid/liquid ratio in the slurry is modest. The effect of retention time and energy consumption was evaluated using the relatively low-carbon Finnish supply mix, however, and the effect of energy is presumed to be more significant in more fossil-fuel intensive countries.

The current study also demonstrated that simulation coupled with LCA can be a powerful tool in evaluating prospective future process options with a limited amount of experimental data. The results can be used to assess challenges in the new process option and to uncover data gaps that require further investigation. The model can also be updated to obtain more accurate results of the environmental impact categories when more experimental data is collected.

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Supplementary information

Supplemental data for this article can be accessed at www.tandfonline/gmpr.

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