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Coal-Gold Agglomeration: An Alternative Separation Process in Gold Recovery

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Abstract: Considering the increasing environmental concerns and the potential for small gold deposits to be exploited in the future, the uses of environmentally friendly processes are essential. Recent developments point to the potential for greatly increased plant performance through a separation process that combines the cyanide and flotation processes. In addition, this kind of alternative treatment processes to the traditional gold recovery processes may reduce the environmental risks of present small-scale gold mining. Gold recovery processes that applied to different types of gold bearing ore deposits show that the type of deposits plays an important role for the selection of mineral processing technologies in the production of gold and other precious metals. In the last 25 years, different alternative processes have been investigated on gold deposits located in areas where environmental issues are a great concern. In 1988, gold particles were first recovered by successful pilot trial of coal-gold agglomeration (CGA) process in Australia. The current paper reviews the importance of CGA in the production of gold ore and identifies areas for further development work.

Keywords: Gold ores, agglomeration, coal, environmental, fine particle processing

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INTRODUCTION

There are several gold recovery processes that are well proven to be able to produce gold on an economical profitable way. Oil agglomeration is one of these alternative processes, which was applied as soon as 1920. In the 80s, it was developed for laboratory scale and pilot scale. The process has been applied successfully on gold recovery in some pilot plants (1, 2). Agglomeration is based on spherical agglomeration or oil agglomeration. The principle of spherical agglomeration has been successfully used for the recovery of various minerals. Spherical agglomeration is a wet process in which size enlargement occurs among particles suspended in liquid phase (3). Oil agglomeration has been applied for fractionation of tin-containing ore (4), antimony ore (5, 6) as well as for the beneficiation of gold ore and high ash content coal (7–26). These are few examples of the selective agglomeration. The gold mining industry has relied mainly upon the use of highly pollutant chemicals, such as liquid mercury metal and cyanide solutions to recover gold from its ores. The advantage of gold recovery by the coal-gold agglomeration (CGA) process has little or no negative impact on the environment. A gold ore containing liberated gold particles is contacted with coal oil agglomerates, whereby gold is then recovered into the coal/oil phase (27).

The use of cyanidation and mercury amalgamation will continue in the future although more stringent and hence more costly precautions to protect both the environment and the workforce can be expected. The commercialization of a novel process to recover gold from certain types of auriferous deposits which will eliminate or at the very least, minimize the use of these potentially hazardous chemicals, will be very positive. The CGA process offers this opportunity. However, not all gold bearing deposits can be economically treated by CGA. A wide ranging program of laboratory test work on various auriferous materials has shown that CGA is most applicable in situations where the gold metal is free, or is readily liberated. In these circumstances gold recovery is found to be independent of particle size. The size of particles that can be recovered ranges from less than 5 microns to plus 300 microns. CGA cannot be used if gold is associated with significant quantities of pyrite, arsenopyrite or other sulfide minerals. Sulfides are as lipophilic as gold and therefore these minerals penetrate readily into the coal-oil agglomerates (28).

Although this new free gold recovery process for small-scale gold mining community is not considered novel, it needs to be optimized especially for the separation stages. It, thus, becomes clear that an effective CGA process that will be capable of operating on a fairly large scale is needed. Flotation was considered as the best process route due to its wide range of applications and ease of operation and implementation.

The flotation process is the most effective, complex and commonly applicable process that is used for ore enrichment in the mining industry. It can be applied to any mixture of particles fine enough to be carried up by rising air bubbles (25).

In the recent years, joint papers on the main international journals and congresses have been produced to publish the results for the development of environmentally friendly and cost effective processes. This paper presents a review of the use of CGA process at gold mining operations on a worldwide basis.

BRIEF REVIEW OF SEPARATION PROCESSES

Recently, it has been established that approximately 13% of gold recovery is realized by physical and 87% by chemical and biological processes (29–34). In most cases it is anticipated that auriferous ash would be treated by leaching in order to recover the contained gold at the very high extraction efficiencies i.e., greater than 99%. The extracted gold would be finally recovered from the leach solution by cementation, precipitation or electro-winning. The leaching product would be thiourea or thiosulfate for applications due to environmental reasons and cyanide would be used. Each application would be judged on its own merits and the most appropriate ash treatment option selected. It should be noted that the final gold production stage is a very small scale batch operation and the quantity of leachants that would be used would be very small compared with a conventional gold cyanidation process (28). Chronological list of gold recovery methods is given in Table 1.

In some gold recovery plants, froth flotation process is applied and combined with amalgamation and cyanidation methods. Gold ore is

Table 1. Chronological list of selected gold recovery methods

Date	Invention/Application
1000 BC	Amalgamation
750 BC	Dissolution of gold by mercury
1783	Dissolution of gold by cyanide salts - Scheele (Swiss)
1889	Foundation of first cyanidation plant - Crown Mine (N.Zealand)
1949	First application in carbon in pulp (CIP)
1970	Applications in heap leaching - Carlin (USA)
1976	Separation of carbon in gold recovery by alcohol
1986	First patent in gold recovery by coal-oil agglomeration
1986	First commercial application of bio-oxidation in sulphide ores - Fairview (South Africa)
1996	First pilot-scale application of thiosulfate leaching in gold recovery - Carlin (USA)

preconcentrated by flotation method. However, flotation has recently been applied due to the environmental problems. Froth flotation has been used for epithermal gold deposits and fine gold particles. The following is used for gold recovery method:

- Disposal of impurities prior to cyanidation
- Recovery of tellurides
- Evaluation of the gravity and the amalgamation tailings
- Evaluation of post leaching of the tailings
- Production of high - grade preconcentrations
- Alternative method for the environment (pretreatment)

Flotation is employed especially for recovery of primary gold ore. It is necessary to investigate economical and technical application of the flotation method. First of all, flotation is commonly applied for the preconcentration products and the tailings in post leaching treatments. Flotation is performed in mechanical and pneumatic flotation machines. Especially, the use of pneumatic flotation machines results in definitely lower investment and energy costs. The use of columns for the flotation of gold ore is likely to present problems since the major objective is a high recovery factor. In the flotation process, optimum reagent addition, optimum pH and optimum flotation time are investigated by tests prior to other gold recovery methods.

Gold extraction involving flotation can be separated into four categories, free milling gold ores, copper gold ores, refractory gold ores and partially refractory gold ores (35). In this time, many novel applications and fundamental studies have been produced to gain a more thorough understanding of the chemical and physical phenomena that occur within the solid-water pulp matrix. However, the flotation behavior of gold bearing minerals, especially free gold particles and refractory gold bearing sulfides remains largely unclear (36). The fine gold fraction is considered relatively easy to float. The natural floatability of gold is enhanced with the use of collectors. The most commonly used collectors for the flotation of native gold, other gold minerals, and auriferous sulfides are the xanthates, typified by sodium ethyl xanthate ($C_2H_5OCSSNa$) (37). The operating pH for the flotation of gold is dependent on the collector (35). In general it was suggested that free gold floats well in the presence of xanthate collectors and froths but not if its particle size is too great or if reagents such as CaO or sodium sulfide are added to the pulp. It was found that activation with copper sulfate did not increase the recovery of free gold but increase the rate of recovery (36, 38, 39). Taggart (39) provided further evidence that the flotation of native gold particles can be affected by physical constraints such as pulp viscosity and the condition of the gold particle surface. He states clearly that native gold is made more difficult to float when it is coated, and also when very fine grinding imbeds other constituents into the surface of the gold particles so that their hydrophobicity and flotation response are reduced. Also, the flotation response of free gold is best in pulps of high-density (36). Refractory gold bearing constituents have long been recognized as being associated with the sulfide component of a particular ore (40, 41). In general, the recovery of refractory gold follows the same trend as the sulfide minerals it is locked in, i.e., pyrite, pyrrhotite and arsenopyrite. Allison et al. (42) found that the use of amine collectors instead of xanthates could be effective for the recovery of pyrite and gold in alkaline flotation circuits. O'Connor et al. (41) found that the recovery of a refractory gold bearing sulfide would enhance gold recoveries by investigating the flotation of refractory arsenopyrite using mixtures of dithiophosphate and dithiocarbamate.

THE COAL-GOLD AGGLOMERATION (CGA) PROCESS

The Historical Development of the CGA Process

The coal-gold agglomeration process for gold recovery became known to the mining industry in the mid-80s after the first related patent was approved. Although the use of oil to recover gold was already introduced in the early 1900s, it is only this late that the potential of oil-coal agglomerates is being investigated as one of the alternatives to the conventional gold recovery processes. Twenty two years have passed since that first patent was issued and various tests and researches had been conducted but the process has not gained its commercial success so far.

The utilization of coal-oil agglomerates in the recovery of gold is based on the natural hydrophobicity/lipophilicity of gold, a property that according to the consensus of most surface chemistry experts is brought about by the ease by which gold surface becomes contaminated. Though it is possible to recover gold by agglomerating them with oil, the amount of gold in the ore is usually so small that there are insufficient gold particles to form agglomerates. Thus, the need to use other hydrophobic materials (e.g., coal) to either form agglomerates together with gold or act as carrier of gold particles was evidenced (21).

Chronological development of oil agglomeration is given in Table 2.

From 1920 until now, many researches has contributed to the enrichment of gold with coal-oil agglomeration techniques. In the last two decades (after the research of BP), coal-oil agglomeration has been investigated in detail by many researchers. Many investigations on this subject have uncovered the importance of coal-gold agglomeration in technical, economical and environmental aspects.

Table 2. Chronological development of oil agglomeration

Process/ Inventor	Year	Feature/Study	Objective
Trent	1920	Selected recovery of coal fines by using oil (30% wt. of dry coal), separation by screening.	Recovery of coal
Convertol	1952	Reduced oil consumption over Trent Process (3–15% wt.%). Mixing of oil slurry in a special phase, inversion mill, final product in floc form, dewatering in solid bowl centrifuge, solids density in coal slurry very high.	Recovery of coal
Reerink, Muschenborn, Notzold	1956	Novel mixing device	Recovery of coal
Mondria (Shell)	1959	Emulsion of light hydrocarbons in water to remove soot particles from aqueous slurry. Claimed to work better than the processes using unemulsified hydrocarbon.	Removel of soot particles from aqueous slurry
NRCC	1960	Low % solids in the slurry, controlled agitation to control the product size, low residual moisture; oil agglomeration step followed by balling drums, discs for pelletization.	Recovery, cleanning, dewatering and pelletization
Kaiser	1964	Use of hydrocarbons to remove carbon particles from waste water. Agglomerate product removed centrifugally. Product in the form of paste with 20–50% water.	Cleaning of plant waste waters
McDonald	1966	Used 200 micron size oil droplets (produced from spray nozzles) to remove contaminations from water.	Cleaning of plant waste waters.
Shell	1970	Shell pelletizing separation-novel design with multistage effect, very low to 30% solids in the slurry.	Cleaning of water
Walsh, Whalley, Botham	1972	Hot suspension of coal and water to allow the use of heavy oils. Produced pellets to be used as feed to coke ovens.	Recovery of coal
Olifloc	1973	Treats very fine and high ash content coal slurry obtained after desliming operations, uses high energy mixers for oil agglomeration, employs flotation for recovering the last bits of carbonaceous material.	Cleaning and recovery of coal

(*continued*)

Table 2. Continued

Process/Inventor	Year	Feature/Study	Objective
Schubert	1974	Pre-emulsification of oil prior to its mixing with coal fines slurry.	Recovery of coal
Shell	1976	New non-selective process for dewatering of pipe-line coal slurries.	Dewatering
CFRI	1976	Solids content in slurry 4–15%, tests done with not easily washable coals.	Recovery and dewatering
BHP	1976	Fundamental work, pre-emulsification of heavy oil before addition to coal slurry. Uses recovery of oil.	Recovery and dewatering of coal
BP	1983	The use of spherical agglomeration for coal and minerals, using the same hydrocarbon bridging liquid, the coal acts as a "carrier" for minerals.	Recovery of coal and fine gold particles
Cadzow, Elkes, Ewin, Mainwaring	1986	First patent for recovey of gold by coal agglomeration.	Recovery of gold
House, Townsend, Veal	1988	CGA method in pilot plant-Australia (~ 1 g/t, 1 tph)	Recovery of gold

An environmental audit has been undertaken on the CGA process. In common with most metal processing operations it identifies tailings disposal as the one area most likely to have the biggest environmental impact. As distinct from conventional gold recovery plants where the tailings can contain appreciable quantities of free cyanide and cyanide complexes, CGA tailings will contain only small quantities of contaminants, namely coal fines, oil, collector (xanthates) and possibly lime. The concentrations of these contaminants will be well below threshold levels so the impoundment of CGA tailings in a tailings dam is unlikely to cause unacceptable environmental problems (28). The barren tailings effluent from a CGA plant can be considered as being similar to that resulting from a base metal flotation plant. Further, as the contact residence time required for CGA is low, so the tank storage is much less than would be required for a cyanidation plant. Additionally, gold is recovered from the feed in a single stage irrespective of gold size distribution rather than in the several stages necessary for ores with a wide gold size distribution if gravity separation is adopted. Consequently, for the same tonnage put through a typical CGA process plant will be smaller in size and therefore less visually intrusive than conventional gold processing plants using either cyanidation or gravity concentration. Much development work has been undertaken on CGA and this has established that the process is viable both technically and economically. The technology will find applications in circumstances when the gold mineralogy of a deposit is favorable and when other metallurgical methods are either unable to produce better gold recoveries or they can not be used because of other constraints.

In most applications cyanide dissolution will invariably give a higher recovery of gold than CGA, even on materials most suited to CGA processing. The better recoveries from cyanidation will therefore inevitably justify the higher capital costs of a cyanide leach system over a CGA plant. However, situations can arise where cyanidation can not be considered at all because of other constraints particularly environmental concerns. In these circumstances CGA represents a realistic and acceptable option to the exploitation of the deposit which otherwise would remain undeveloped. Recently, attention has been focused on gold exploration activities in Western Ireland, in the mountains and off-shore islands of County Mayo and Connemara. Promising results have been obtained by exploration companies. However, exploitation of these deposits is being actively resisted by a strong environmental lobby. Fears concerning the pollution problems experienced at other recently closed mines in Ireland, particularly involving the use of cyanide, will inevitably preclude the use of cyanidation should these deposits be developed. The only environmentally acceptable alternatives that would be viable for this location are therefore gravity concentration and CGA. Both processes would require liberation of the gold prior to concentration to be effective. If the gold is in a narrow, relatively coarse size range where one stage gravity concentration can be employed to give acceptable recoveries then gravity concentration can be used as well as CGA. If, however, the gold is in a wide size range where several stages of gravity concentration are required and where some gold particles are too fine to be recovered by gravity, then CGA is the most economically appropriate technology (28).

Performances of the CGA Process

Since the early 1980s, many researchers in several countries have been experimenting with the CGA process with various types of gold bearing ores. It has been proved that the CGA process is of extensive suitability to a variety of gold ores.

Placer, alluvial ores and beach sands: The gold particles in these gold bearing materials are mostly free from gangue, hence, grinding can be omitted or attrition scrubbing may be needed to liberate cemented gold. In most case, the coarse gold particles may be firstly recovered by gravitational concentration, but recovering fine gold particles is beyond the reach of this method. Bellamy et al. (12) claimed that fine gold particles (<5 μm) and relatively coarse gold particles (ca. 500 μm) were recovered by CGA with equal ease. They tested six different samples of placer sands from the different deposit sites in New Zealand, which had significant variations in their compositions and surface properties. It was demonstrated that there existed an optimum grinding size in either dry grinding or wet grinding; as a matter of fact, attrition scrubbing could make gold free from cemented pellets; and the addition of collector (KAX) and higher agitating rates could greatly improve the gold recoveries. Therefore, the desirable flow sheet involved attrition scrubbing, size fractioning and processing the undersize with CGA. The batch tests, which treated head feed grade 0.21–0.78 ppm, could get gold recoveries of 85–98%, with the gold grade in the tailings as low as 0.03–0.09 ppm. Beltran and Jacinto (43) tested alluvial tailings containing Au 3 g/t, and got the concentrate containing gold in excess of 900 g/t at recoveries of about 80% by using coal dust and diesel oil as agglomerating agents, flotation for separating loaded agglomerates from the slurry and the consumption of coal and oil was about 20 kg/ton and 2 kg/ton tailings respectively. Cadzow and Lamb (44) conducted alluvial ore tests from batch cell (1 liter) to 5 t/h semi-commercial trial (in CARBAD plant 3×1 m^3 tanks). The ore fluctuated in gold grade between Au 0.56 g/t and 0.105 g/t, but recoveries of up to 90% were achieved. Moreover, the laboratory experiments with alluvial ores, clays and beach sands from Australia and New Zealand were able to get gold recoveries of 91–99%.

Gravity tailings: The BP's researchers (2, 45–47) conducted different scale tests from laboratory batch test, to continuous pilot trial (1t/h) on the 'sludges' in Australia. The sludges, which had been placers and hard rock ores in origin, resulted from decantation methods used for gold in the early 20th century. In the pilot trial, 3 types of ore samples (A, B and C) from different deposit sites were tested. The sample A mainly consisted of quartz, kaolinite, muscovite, serpentine, goethite and limonite, and no sulfides were detected (<0.2%). The mean gold head grade in the sample A was 0.56 g/t (the determined grade range was 0.38–0.81 g/t). The gold present in the sample A, 20% was naturally lipophilic, 40% recoverable by addition of a collector and 20% locked to gangue but could be liberated by grinding to $D_{80} = 70\,\mu m$. Maximum gold recovery with grinding was therefore 80%. In the pilot trial, the highest recoveries were about 75%, resulting in tailings grade of 0.15 g/t.

Leaching residues: Zhao and Wang (48) and Zhao and Lu (49) treated a type of heap leaching residue, which contained Au 4–6 g/t, some lime and remainder of cyanide. The original ore had been quartz-oxide type. The gold existed in free form or associated with pyrite and arsenopyrite. The continuous trial (1 t/d) indicated that for the head residue grade of 5.9 g/t Au, the tailings contained Au 1.01–1.20 g/t. The content of gold in the agglomerates was 456 g/t.

Amalgamation residues: Zhao and Lu (50) and Zhao and Lu (49, 51) performed tests on an amalgamation residue containing Au 5.04 g/t and Ag 84 g/t. Its main metal minerals were limonite, hematite, jarosite, gold and electrum, as well as a small amount of galena, pyrite and chalcopyrite and its main non-metal minerals were calcite, quartz and feldspar. The gold was closely associated with gangue and was of fine sizes ranging from 0.01 to 0.053 mm. Such parameters as grinding size, pH value, collector usage, agglomerate usage and agitation rate were tested. The optimum condition: Grinding size 95% −200 mesh, slurry pH 9.0, combined usage of xanthanate (150 g/t) and aero float (100 g/t), agglomerate usage 250 kg/t, agitating rate 800 rpm. An adsorption recovery of 88% was achieved, and the tailings contained Au 0.6 g/t. Moreover, the combustion, chlorination leaching and smelting following the CGA adsorption were tested. Finally, the final gold bullion product contained Au 99% and the overall recovery of the whole process was over 85%.

Hard rock ores: Kilinc (17) and Akar et al. (19) tested a gold-in-quartz ore excavated from epithermal deposit and its average grade was Au 10–11 g/t, and the liberation size of gold particles was between 10 and 40 microns. Under the optimum conditions of laboratory test, the recovery and concentrate grade were 75.2% and Au 51.5 g/t respectively, and the tailings grade was Au 3.36 g/t. Compared to flotation, the more quality concentrate product might be obtained. Sen et al. (52, 53) tested some experiments to reveal the reaction of the coal oil assisted gold flotation process against size and the number of gold particles in the feed. The effect of changing coal quantity for constant ore and oil amounts was also investigated. They observed that there is no significant difference in process gold recoveries for feeds assaying different Au. They found that the process seems to be effective for the recovery of gold grains as coarse as 300 μm. The experiments showed that the process gives very similar results for both artificial and natural ore samples; the best results have been obtained by using 30/1 coal-oil ratio. Cadzow et al. (1) tested a series of hard-rock ores and found that their recoveries were somehow affected by grinding size, for example the gold recoveries of a copper-rich super gene ore in North Queensland increased from ca. 69 to 87% as a function of gold liberation when grinding sizes varied from ca.55% −63 μm to 85% −63 μm. Some trial plant modifications enabled the good correlation between the laboratory and semi-commercial plant (5 t/hr), for a hard rock ore, the laboratory batch test yielded gold recovery of 86%, and for the same ore the plant trial recorded a recovery of 85%.

Zhao and Lu (50, 54) tested a gold bearing ore which was an oxidized type and contained many slimes and iron oxide minerals as limonite, hematite and goethite. The ore contained Au 17.5 g/t, Ag 75 g/t and Fe 22.1%. The gold was mostly distributed in size range of 0.005 to 0.04 mm and closely associated with limonite. The previous tests including amalgamation, cyanidation and flotation indicated that this ore was a refractory gold ore. The test made many efforts on cleaning and removal of iron before contacting with agglomerates. It was believed that before adsorption desliming by flotation and removal of iron oxides with diluted HCl solution should be adopted to get an acceptable CGA recovery. Yang and Wang (55) tested various types of gold ores by CGA, and compared the adsorption results with cyanidation leaching. It was shown that CGA was of an extensive suitability for various gold types, including oxidized, carbonaceous, carbonate, and erosive rock containing gold, and got excellent recoveries which were mostly 2 to 3 points (%) higher than cyanidation leaching for the same ore, and the enrichment ratios in a single adsorption generally ranged from 36 to 58 times. It was also proved that among the tested ores CGA was particularly suitable for the slimable and oxidized gold bearing ore, and for high content of sulfides, the adsorption recovery were very high, but the enrichment ratios were too low. Buckley (56) recommended the order of amenability to CGA process of several gold ores (from good to bad): gravity concentrate, free milling, placer, flotation concentrate (oxidized type), gravity tailings, oxidized zone ore, pyrite ore and no-gravity tailings.

Based on the papers in the literature, the responses of CGA process to various types of gold ores were briefed in Table 3.

The Mechanism of CGA Process

The coal-oil-gold agglomeration process considers the preferential wetting of coal and gold particles. It takes advantage of the greater hydrophobicity and lipophilicity of coal and gold compared to that of most gangue materials (53). A schematic diagram of the agglomeration steps is shown in Figure 1.

The CGA process involves the selective recovery of lipophilic gold particles into the coal-oil agglomerates from aqueous slurry. The coal-oil agglomerates are pre-formed by intensive agitation of an aqueous coal slurry and oil. Intensive agitation provides the distrubition of oil into water in fine spherical droplets and the collision of coal particles

Table 3. The responses of CGA process to various gold ores

Gold ore type	Head grade (Au g/t)	Tailings grade (Au g/t)	Gold recovery (%)	Remarks	Reference
Gravity concentrate (from centrifuging)	980	85	90.5	Batch test, just one cycle	(15)
Old gravity tailings (from slucing, no suphides)	0.56	0.15–0.19	62–75	Pilot trial	(2, 45, 46, 47)
Alluvial ore	0.105–0.56		90	Semi-commercial. CARBAD media.	(44)
Heap leached residues (quartz-oxidized type)	5.9	1.01–1.20	Ca.80	Continuous trial. ground to −200 mesh. 85%	(48)
Amalgamation residue	5.04	0.6	88	Batch test. The ash containing Au 3042 g/t.	(50)
Oxidized gold ore	16.0	2.42–2.82	82.4–85	Continuous trial.	(49)
Arsenopyrite-pyrite-quartz	6.11		93.3	Batch test.	(44)
Limonite-pyrite-quartz	2.4		95.5		(44)
Pyrite-quartz	1.48		79.7		(44)
Chalcopyrite-pyrite-quartz	14.1		88.6		(44)

Flotation concentrate (oxidized type)			60–85	(56)
Flotation concentrate (sulphide type)			0–10	(56)
Telluride (over 10% gold presented in telluride form)			82–94	(56)
Quartz-oxidized ores	3.0–18.5	0.18–0.79	94–95.7	(55)
Slimable oxidized	13.6	<0.1	>99	(55)
Carbonate	2.8–5.1	0.1–0.25	96.4–97	(55)
Errosive rock	5.73–6.74	0.24–0.31	Ca.96	(55)

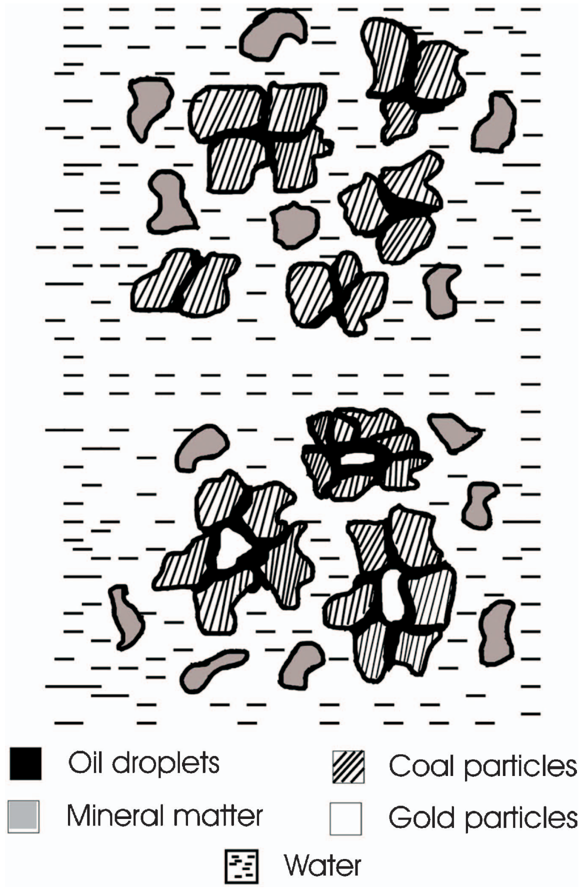

Figure 1. Schematic diagramme of agglomeration steps.

and fine oil droplets (24, 57). Upon agitation and subsequent collision (Figs. 2 and 3), the oil droplets overspread on the coal surfaces and the collision of these oil wetted particles causes formation of "oil bridges" (10, 15, 24, 58). In this way, coal particles are converted to the agglomerates which are combined by oil bridges. The coal-oil ratio, oil type and agitation intensity determine the size of the agglomerates, which are typically 2 mm in diameter. Gold recovery is achieved by contacting the pre-formed agglomerates and gold slurry under conditions of intense agitation. The gold particles, which are substantially free, become attached to the coal-oil agglomerates as they collide and eventually the gold penetrates into the agglomerates. Gold loadings of 5 to over 10 kg Au per ton agglomerates can be achieved with this CGA process (28).

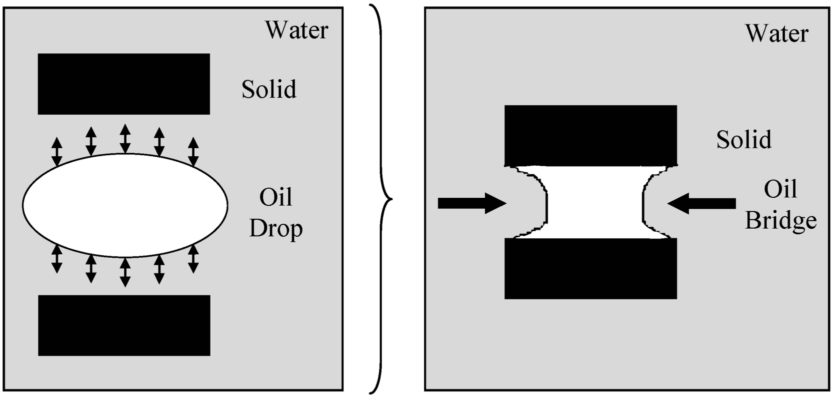

Figure 2. Surface tension in oil agglomeration.

The effect of various parameters on CGA process

It is possible to gather the effective parameters of CGA in two main groups. These parameters can be defined as material parameters and operational parameters.

Dispersed gold bearing ore in slurry, carbonaceous material and oil phases are determined in material parameters. The chemical, physical and physicochemical properties of these phases effect the achievement of the CGA process. Especially, mineralogy of gold, type, amount and size of carbonaceous material, type and amount of oil are important parameters for this process.

In the CGA process, gold grains ranging from 1 to 500 microns can be recovered with equal ease (25). The process is most suitable for treating alluvial or free milling ores, gravity concentrates and current or old gravity plant tailings (12, 25). If the gold ore involves pyrite, arsenopyrite or other sulfide minerals, CGA may not be used successfully. Sulfides are as lipophilic as gold and would be readily

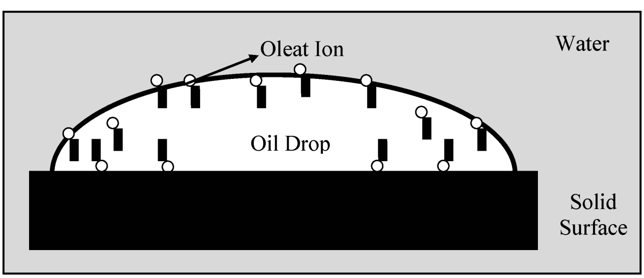

Figure 3. The spreading of oil on a solid surface.

absorbed into the coal-oil agglomerates (28). It has been reported that there is no significant difference in gold recoveries with up to 5% sulfides in the feed (21, 53).

Graphite, bituminous coal, industrial coal, metallurgical coal, lignite coal (low ash content) has been used successfully by many researchers in the CGA process. The successes of coal-oil agglomeration are determined by surface properties of coal. The surface properties of coal depend strongly on three parameters. These are a) the hydrocarbon structure depending on the rank of coal, b) the type and number of polar groups containing especially oxygenated functional groups, c) The content of inorganic materials in coal (59, 60).

Crude petroleum, various distilled petroleum fractions such as diesel oil, pure aliphatic hydrocarbons and vegetables oils were used as agglomerating agents for oil agglomeration of coal in CGA process (11, 17, 21, 53, 57, 61). Furthermore, the gold adhesion process is significantly improved in the presence of collectors such as xanthate in CGA (2, 15, 62). For xanthate collectors with different hydrocarbon chain lengths, the longer the chain, the more efficient is the attachment of gold to the agglomerates (62).

In the CGA process, there are operational parameters that affect the outcome of agglomeration and gold recovery. These operational parameters affect the sub-processes which are the dispersion, collision and adhesion of the phases in the pulp (24). With adequate amounts of oil and sufficient mechanical agitation, oil-coated particles collide with each other and form agglomerates due to the interfacial tension of the oil and the capillary attraction of the oil bridges between particles (10, 21). Mechanical agitation increases oil dispersion and the collision of coal particles with oil droplets. Furthermore, increasing pulp temperature or adding surfactant in pulp plays an important role in the emulsification of oil (63–66). A general flow sheet of coal-oil agglomeration has been presented in Figure 4.

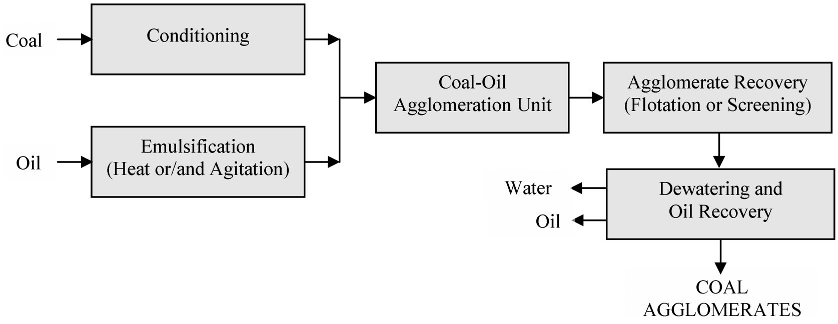

Figure 4. A generalized process flowsheet for coal-oil agglomeration.

Gold recovery into the agglomerates is not limited by loading. Therefore, recovery kinetics is constant with time and independent of head grade. High selectivity can be achieved against gangue minerals by reagent control, as in froth flotation (28). The recovery of gold by CGA process depends on these factors:

(a) degree of inter-particle collisions
(b) agglomerate strength and stability
(c) type, degree and time of agitation
(d) contact time and number of recycling of the agglomerates (15, 21, 25, 62).

Agglomerates may be produced in a two stage process. In the first stage, an lipophilic material with (30–70%) is added under high shear conditions to form the micro agglomerates and for the second step, the rest of the material is added under low shear conditions to increase the size of the agglomerates (15, 16). The coal-oil-gold agglomeration process consists of two separate parts: production of coal-oil-gold agglomerates and recovery of these agglomerates from pulp. Flotation of the agglomerates or screening can be employed for the recovery stage (2, 12, 15, 26, 47). A general flow sheet of coal-oil agglomeration has been presented in Figure 5.

CGA process involves either the attachment of gold to oil-agglomerated coal (Fig. 6) or the simultaneous agglomeration of gold with coal and oil (Fig. 7) (21). The pre-agglomeration stage (Fig. 6) increased the number of coal agglomerates in the slurry and consequently, the agglomerates surface areas for gold penetration. Increasing the number of agglomerates in pulp and providing enough surface area for gold particles penetration gave rise to gold recoveries (53).

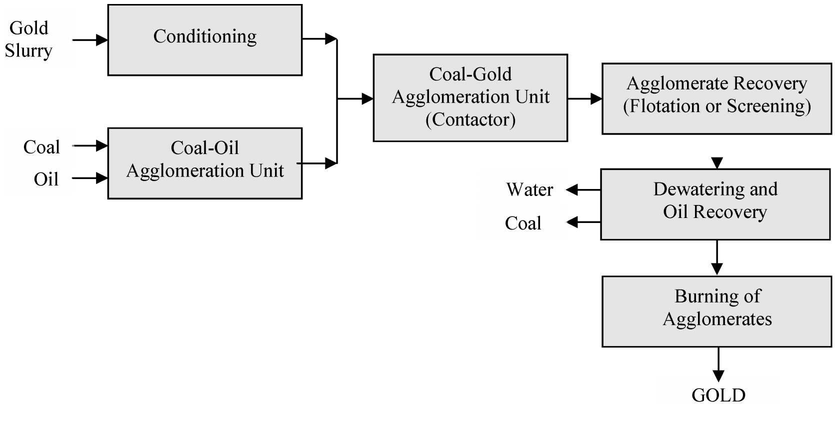

Figure 5. A general flowsheet of coal-oil-gold agglomeration process.

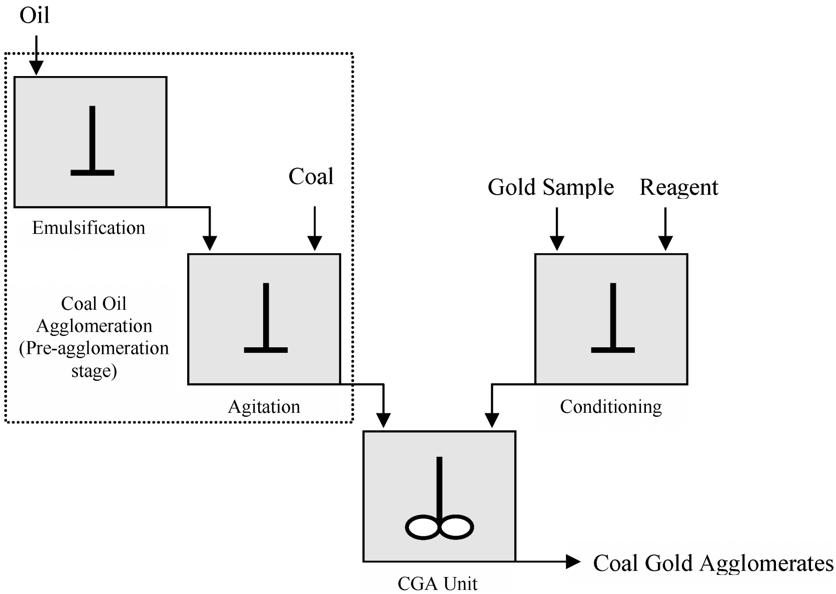

Figure 6. Schematic diagrammes of production of coal-oil-gold agglomerates with pre-agglomeration stage.

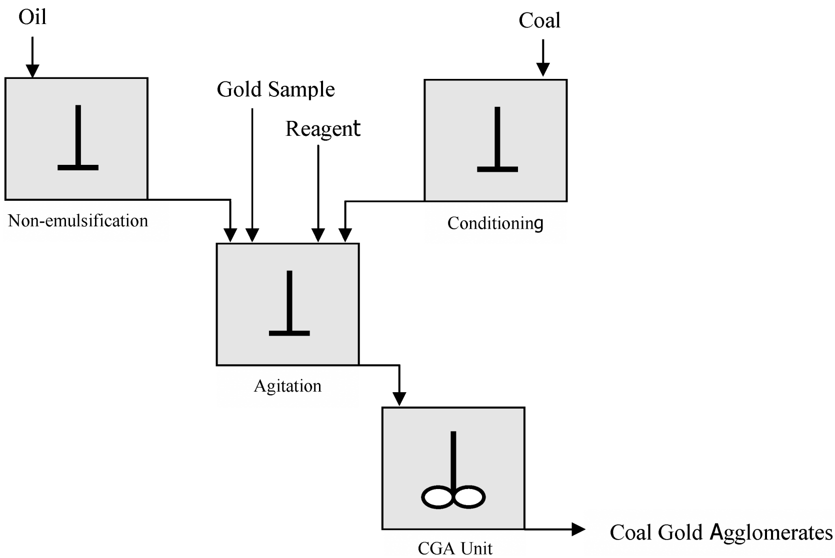

Figure 7. Schematic diagrammes of production of coal-oil-gold agglomerates without pre-agglomeration stage.

Without pre-agglomeration stage (Fig. 7), production of coal-oil-gold agglomerates in ore slurry was not successful (53). As tested before, saturated hydrocarbons (oil) which are insoluble in water need to be dispersed into fine droplets to interact with the coal particles. Decreasing the size of droplets will increase the number of these droplets in the cell and therefore increase the collision between oil droplets and coal particles (24). Hence, only a limited number of agglomerates could be generated, about 25% of coal particles were converted to coal agglomerates (53).

Calvez et al. (21) and Wu et al. (62, 67) indicated that the coal agglomerates surface area for gold particles penetration and the number of coal agglomerates for inter-particle collisions play the major roles in the process. For this reason, increasing the number of gold particles in pulp for the same limited amount of coal agglomerates caused recovery losses in the experiment.

Fundamental Aspects of Coal-Gold Agglomeration

Parameters affecting the coal gold agglomeration process which are identified as material and operational parameters have been investigated by many researchers. The influences of agglomerating agents containing carbonaceous hydrophobic materials and hydrophobic oils on CGA are summarized in Table 4.

Although Table 3 lists the adaptability of CGA to various gold ores and exhibits the variations of CGA performances but from the viewpoint of mineralogy, some common rules can be derived as in Table 5.

The responses of CGA to operational parameters are presented in Table 6. The general operational parameters of CGA process include conditioning, recycling, agglomerates/ore, slurry pH, contacting time, etc. It is very difficult to define a precise and the most suitable value for any parameter.

Several studies were undertaken to evaluate the merits of this process. Bellamy et al. (12) studied the effects of attraction scrubbing, wet and dry grinding and collector (potassium amyl xanthate) addition on gold recovery from beach sand deposits. They also compared the CGA process to conventional gold recovery techniques such as gravity concentration, cyanidation and flotation by using samples from the same deposit and found the CGA process to be quicker, cleaner and more effective (25).

Cadzow and Lamb (44) presented results on the applications of the process as a primary recovery method for hard rock and alluvial ores, for gravity and flotation concentrate upgrade, and for tailings re-treatment. House et al. (14) obtained encouraging results from experiments conducted on four gold bearing materials: synthetic silica/gold mixture,

Table 4. The influences of agglomerants on CGA

Carbonaceous material	Hydrophobic oil	Remarks	Reference
Coal, $d_{80}=100\,\mu m$, ash 18.5%.	commercial grade gas oil density 840 kg/m^3	batch test, several gold ores were tested. under optimum conditions, over 95% recoveries obtained.	(46)
Coal ash 8%, $d_{80}=90\,\mu m$	diesel fuel, density 835 kg/m^3	pilot trial. Over long recycling period, the loading increased constantly with recycle time.	(2, 45, 47)
Carbon	hydrocarbon	semi-commercial trial. The agglomerates 3 mm and stable.	(44)
Seam coal 90% $-100\,\mu m$	Kerosene, fuel oil or light gas oil	light gas oil was better than the two oils. not pre-forming agglomerates.	(1)
Coal dust	diesel oil	the consumption of coal and oil was 20 and 2 kg/t, respectively.	(43)
Coal (10–500 μm)	gas oil	forming agglomerates in two steps.	(69)
Graphite, coal (16.7% ash, 51.4% fixed carbon, ground to 100 -0.42 mm and 36% $-37\,\mu m$), charcoal.	kerosene, diesel oil, soya oil, corn oil	synthetic graphite agglomerated Very well, coal well, charcoal badly. Diesel oil better than the others.	(15)
Graphite 100% -0.2 mm	olive oil, soya oil	the viscosity and recovery of olive were higher than those of soya oil.	(17, 19)
Black coal, two types of lignites, ground to 100% -0.56 mm, and the slime fraction of $-10\,\mu m$ rejected.	olive, kerosene, pine oil. used as an emulsion with distilled water	black coal better than the lignites. olive better than the others. Oil/Coal 0.25.	(16)
Bituminous coal (54–74% fixed Carbon, 10–19% ash, $-300\,\mu m$)	diesel (840 gpl), kerosene (790 gpl)	diesel better. Oil:Coal 0.37 and finer coal got higher recovery.	(21)

(*continued*)

Table 4. Continued

Carbonaceous material	Hydrophobic oil	Remarks	Reference
Coal: low ash, high volatile, −90 to −120 mesh.	diesel oil (containing Surfactants)	coal :oil 100:15–25, agglomerate size −30 to −60 mesh.	(70)
Coal: low ash, < 38 μm.	Creosote + tetralin-red Crude oil	coal : oil 1: 1, agglomerate size−0.5 mm. Suitable for recovering Au loaded activated carbon in carbon in pulp/carbon in leach processes	(71)

black sand gravity concentrate from Indonesia, gold tailings from France and tailings sample from Australia. Marciano et al. (15) investigated the effect of the type of coal, type of oil, the coal/oil ratio (w/w) and agglomerate: ore ratio (w/w) on the performance of the CGA process. Other operating parameters that were also investigated include the contact time and the recycling of the agglomerates, up to six times. They found that the amalgamation and CGA experiments yielded similar results.

Gaigarjiev et al. (16) observed the adhesion of gold particles on the surface of agglomerates by taking digital photos of gold-loaded agglomerates. Calvez et al. (21) examined the relationship between

Table 5. The effects of mineralogy of gold bearing ores on CGA

Mineralogical factor	Effects on CGA
Gold head grade	with the increase of gold grade from 0.5 to 30 g/t Au, recovery of gold increased from 60 to 98.8%, but in the range of 30 to 120 g/t. The rule was broken (1).
Gold particle size	the CARBAD process has ability to recover gold particles from sub-micron size up to 400 μm (44). High recovery indepedent of particle size, from fine (minus 5 microns to coarse (plus 300 μm) (2).
Gold inclusion	the gold in a mechanically-coherent, porous cement (47) and in solid solution (1) can not be captured.
Containing sulphides	gold recovery is not affected by the presence of small amount of sulphides (<5%), but it could be significantly affected by a higher amount (21).
Containing iron oxides	iron oxide minerals like limonite, hematite, goethite had adverse effects (54).
Containing heavy sand	the common heavy minerals like rutile, garnet, magnetite, quartz, did not significantly affect gold recovery (16).

Table 6. The responses of CGA to operational parameters

Operating parameter	Remarks
Addition of collector	collector addition range of 0–250 g/t. The benefit of collector addition (generally KAX) is significant for increasing gold recovery (2, 12, 17, 19, 26, 50).
Agglomerate/ore	proper ratios: 1.1 (15), 250 kg/t (50), 0.4 (21), 0.1 (56).
Slurry pH	pH 4–12 (21), pH 8–10 (50).
Agitating rate	high agitating is favourable for higher selectivity (50).
Pulp density	10–50% solids by wt. (56).
Contacting time	gold loading of agglomerates increased nearly at a constant rate (2).
Agglomerate recycle	increasing recycling number increased gold loading stably, but in each cycle, gold recovery decreased (15).
Maximum gold loading	1000 g/t (recycling 230hrs) (2), 1300 g/t (recycling 20 times), 18 kg/t (recycling 6 times, feed grade 980 g/t) (15), 456 g/t (50).
Ashing temperature	about 800°C (15).

agglomerate surface area and ratio of oil to coal, and the effect of available surface area of agglomerates on gold recovery. Furthermore, by examining the surface and cross-section of agglomerates with SEM, it was observed that gold particles were attached at the agglomerate surface as individual grains or as clusters, some particles were embedded between coal particles, and others were buried deep within the agglomerates. Akcil (20) investigated the effect of the type of oil and the coal/oil ratio on a Turkish gold ore for finding an optimum performance of the CGA process. Bouwer (22) performed work on a South Africa coal, with ethyl oleate as an agglomerating agent on a small scale. They succeeded in optimizing the operating parameters for this specific combination of coal and oil. It was however evident that the separation efficiency could still be improved, which in turn would result in much better gold recoveries. The scale on which the experiments were done was impractical in that it would be impossible to use in a small-scale mining environment.

Moses and Petersen's study (25) indicates that increasing the stirring rate to a certain value results in obtaining higher gold and mass recoveries for the flotation of agglomerates. Wu et al. (62) investigated the adhesion behavior of gold particles to coal-oil agglomerates, and revealed that in addition to gold particles, gold flocks and even micro-nuggets can be detected on the agglomerates. The authors say these flocks are probably formed by the movement of initially penetrated gold particles over the surface of the agglomerates.

Sen et al. (53) found that high recovery values could be obtained by coal-oil assisted gold flotation without any recycling stages of coal-oil-gold agglomerates. Relatively high recovery and superior grade values in comparison to the conventional flotation could be obtained by coal-oil assisted gold flotation for these problematic particles. Therefore, increasing the coal/oil ratio gave rise to the recovery, whereas the grade of the concentrates decreased for both synthetic and natural gold ore samples.

The Adhesion Behavior of Gold Particles to Coal-Oil Agglomerates

For an agglomeration system, if the gap between the hydrophobicity of related agglomerating agents (the solid and the liquid) exceeds their tolerances to each other, it is difficult for an agglomeration process to proceed properly, especially to produce appropriately sized dense agglomerates. So, two factors such as hydrophobic compatibility between the solid and the liquid, and agglomeration coefficient were proposed for quantitatively assessing an agglomeration process. The use of some organic surfactants was able to adjust their compatibility and make the agglomeration system work well. This promotion action was explained in terms of the composition of the interfacial interactions and the immersion energy of the system. Namely, the fact that adding organic surfactant (for example, Di-n-butylphthalate (DBP)) into hydrocarbons is able to improve their agglomeration performance is attributed to two factors. Either it decreases the interfacial tension between hydrocarbons and water, and these results in better dispersion of hydrocarbons in water or it increases the wettability of hydrocarbons on graphite powders, and this also results in easier adhesion of oil to graphite particles as well. So-called non-polar graphite surface actually contains some extent of polarity. Adding polar DBP into pure non-polar hydrocarbon oil makes the oil mixture with some polarity. Thus, adding DBP increases the compatibility of oil with the solid in an oil-water-solid agglomeration system, or makes solid particles more easily immerse into oil droplets by increasing polar attractions at the interface of solid/oil. Numerically, the compatibility can be represented by two concepts: the maximum tolerance of solid polarity for immersion, $\gamma_{s,\ \max}{}^{P}$, and the minimum polarity of the corresponding oil for compatibility with the polar solid, $\gamma_{h,\ \min}{}^{P}$.

In addition, the agglomeration comparison among different agglomerating agents can be eventually evaluated by the 'agglomeration coefficient', which is measured as a whole by the capillary rise method, and reflects the physical properties of the liquids and the mutual interactions between the solids and the liquids. Through modeling the agglomeration empirically, the agglomeration performance can be related to the so-called 'agglomeration coefficient'. This coefficient is quite easily worked out, and quantitatively compares the agglomeration behavior of different liquids and solids, and should have practical meanings in predicting agglomeration and selecting agglomerating agents (68).

Based on the theoretical analysis, the adhesion of fine gold particles to oil-carbon agglomerates was quantitatively investigated in terms of adhesion kinetics, rate constant, collision rate, attachment probability and surface energy change. The suitability of the special 'first-order' adhesion kinetics was confirmed, and this kinetics equation involves nearly all common operating parameters (the dosage and size of agglomerates, agitation intensity and contacting time) and makes it possible to predict the influence of all these factors on this adhesion process. The relationship between the attachment probability and the surface energy change can be well described by a proposed 'nucleation model'. The surface energy change of an adhesion process can be calculated by a formula, $\Delta w'_{\max} = \frac{1}{2}\sigma_{hw}(\cos\theta - 1)$, and more importantly it can be empirically correlated to the attachment probability through an exponential equation, $P_a = A^{*}\exp(-\Delta G_{surf}/k)$, called 'nucleation model' as fine gold particles possibly adhere to agglomerates in a way similar to the formation of an 'oil embryo' or 'oil nucleus' on gold surface (67).

The adhesion behavior of gold particles to agglomerates of oil and carbon and the dynamic changes of the agglomerates during the contact process have been investigated. It is shown that the rate of adhesion of gold particles to agglomerates can be significantly increased by using xanthate collectors. It is further demonstrated that the longer the hydrocarbon chain of the xanthate molecule, the greater the rate of adhesion of gold to the agglomerates. The adsorbed gold particles do not markedly affect the adsorption activity of the agglomerates, i.e. recycling the agglomerates to increase the gold loading is feasible. Examination of gold-loaded agglomerates by scanning electron microscopy showed that in addition to individual gold particles, gold "flocks" and even "micronuggets" can be detected on the agglomerates. These flocks and nuggets are probably formed by the movement of initially adsorbed gold particles over the surface of the agglomerates. Both the composition and structure of the agglomerates change during the contact process. The oil tends to diffuse out of the agglomerates, and their oil content decreases with agitation. The gold particles can also migrate into the interior of the agglomerates due to their deformation resulting from agitation impact (62).

SUMMARY

The extraction of gold from the earth has been performed with vigor since it developed an essential value early in the history of mankind due to its unique physical and chemical properties with its decorative appeal and its scarcity. Recently, it has been established that approximately 13% of gold recovery is realized by physical and 87% of that is carried out by chemical and biological processes. Coal gold agglomeration is thought to be an alternative process for gold production in view of technique, cost and environmental friendliness. Although the flotation process is effective and a commonly applicable process, this process used for gold ore enrichment is complex. Gold bearing materials which show consistently high recoveries using CGA are free milling ores, placer ores and tailings from existing gravity concentration plant. In the CGA process, gold recovery is much higher than the flotation process. This is why this process is applied with achievement in small-scale gold mining community. In this process, the cost of oil is the significant portion of the total operating cost. Therefore, a process that provides an oil recovery step will provide an impetus for faster commercialization. Besides, recycling of the agglomerate phase showed that gold loading on the agglomerates could be increased to reduce operating cost.

The coal gold agglomeration process involves the selective recovery of lipophilic gold particles from aqueous slurry into the coal-oil agglomerates. The coal-oil agglomerates are pre-formed by intensive agitation of an aqueous coal slurry and oil. Coal-oil bridges are initially formed and these build-up into flocks and finally into agglomerates. The gold particles, which are free or readily liberated, become attached to the coal-oil agglomerates as they collide and eventually the gold penetrates into the agglomerates. The lipophilicty of gold is enhanced with the use of collectors. The size of gold particles that can be recovered ranges from less than 5 microns to plus 300 microns. CGA cannot be used if the gold is associated with significant quantities of pyrite, arsenopyrite or other sulfide minerals. This method allows these benefits of agglomeration to be obtained on low-grade materials by successive loading and applies particularly to gold where concentrations are very low.

Up to now, coal gold agglomeration has been investigated in detail by many researchers. Coal gold agglomeration process is developed depending on the progress of coal-oil agglomeration. The increasing of higher gold grade and higher gold recovery production demand is the reason for the improvement of CGA method. Being environmentally friendly and an easy to apply method is the biggest advantages that the CGA provides. All research done in terms of operational cost will further increase the feasibility of this method.

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