Introduction

Environmental protection has become the focus of world-wide research in the gold mining industry and favours the development of environmentally sound processes, such as the Coal Gold Agglomeration (CGA) process, which is an alternative to existing gold processing methods, such as cyanidation and mercury amalgamation.

The CGA process was developed and patented by the British Petroleum (BP) research team in the early eighties and is based on the recovery of hydrophobic/oleophilic gold particles from ore slurries into agglomerates formed from coal and oil. The oil acts as the bridging liquid between the coal and gold particles where the coal is the carrier of the mineral and enables effective separation of the oil phase. The coal-oil agglomerates are recycled to increase their gold loading, after which they are separated and further treated to produce gold bullion1-6.

Figure 1 shows a schematic representation of the basic concepts involved in the CGA process.

Gold bearing ores containing liberated/free gold particles, such as alluvial/free milling ores, gravity concentrates or gravity plant tailings was found to be suitable for the CGA process, as associations with other minerals/metals, such as pyrites, arsenopyrite etc. reduce the oleophilicity of gold, and hence limits gold recoveries2,3,6. High gold recoveries, irrespective of the gold grain size (ranging in size from 1–500 µm) were found, therefore providing an alternative to amalgamation or intensive cyanidation3,4,6.

Although the CGA process is not a novel process, sensu stricto, and many further studies besides those done by BP have been conducted7-11, it was attempted in this study to investigate and optimize the process route of the CGA process so as to provide a valid alternative to small-scale mining operations, such as mercury amalgamation.

Theory

A theoretical approach to the CGA process has produced a set of mathematical equations, whereby initial and final conditions were expressed in terms of free energy. Refer to Figure 2, where stage I is a representation of the initial contact of the three main components (gold, water and oil) and stage II is a simplified illustration of the final condition in which the components find themselves after the contact period. The following Equations were used to express the free energy for each stage12:

\[
F_I = A_1 \gamma_{13} + A_2 \gamma_{23} \quad [1]
\]

\[
F_{II} = A_1 \gamma_{12} + A_3 \gamma_{23} \quad [2]
\]
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Equation for the volume of a sphere \[ V = \frac{4}{3} \pi r^3 \]

The resulting Equation was simplified by utilizing the area terms into radii so that the final Equation was found to be as follows:

\[ F_{II-I} = \gamma_{12} (k_2 \theta + 1)^{2/3} \frac{k}{k_2} \cos \theta \]

where:
- \( F_{II-I} \) - the free energy for each stage (J)
- \( A_1, A_2, A_3 \) - the surface areas of each given component (m²)
- \( \gamma_{12}, \gamma_{13}, \gamma_{23} \) - the interfacial tensions for the different components (J/m²)

The above Equations were combined with Young’s Equation):

\[ \gamma_{12} = \gamma_{13} \cos \theta \]

which is a relationship between the various interfacial tensions and three-phase contact angle (\( \theta \)) measured through the oil phase on the gold surface in the presence of water. The resulting Equation was simplified by utilizing the Equation for the volume of a sphere \( V = \frac{4}{3} \pi r^3 \) to convert the area terms into radii so that the final Equation was found to be as follows:

\[ F_{II-I} = \gamma_{23} (k_2 \theta + 1)^{2/3} \frac{k}{k_2} \cos \theta \]

where:
- \( F_{II-I} \) - total free energy difference for the system per unit area (J/m²)
- \( \gamma_{23} \) - interfacial tension between the oil phase and water (J/m²)
- \( k \) - ratio of radii of the agglomerates to gold particles (agglomerate/gold particles)

The total free energy difference of a system \( F_{II-I} \) is a measure of the stability of the system, whereas a lower value represents the more stable system. Equation [4] serves as an empirical model which is a useful tool to evaluate the CGA process in terms of certain parameters such as radii of agglomerates and gold particles or the three-phase contact angle, while \( F_{II-I} \) is non-dimensionalized by dividing both sides of the Equation by \( \gamma_{23} \). A sensitivity analysis on the dimensionless value of \( F_{II-I} \) has shown that the CGA process is favoured by a minimum contact angle \( (\theta) \), where this angle is a measure of the wettability of the gold by the oil phase and must range between 0 and 90° which is classified as the perfect to non-wetting contact angle range. Furthermore, the relationship between the dimensionless free energy and \( k \) was found to be an asymptotic function and has provided information about the size of the gold particles that is recoverable for certain size agglomerates. Furthermore, it was found that the agglomerates had to be larger than the gold particles if these were to be recovered successfully. For the purpose of this paper it was decided not to focus on the model and predictions made from it, but rather to provide the reader with a brief background as to where it originated from and to identify possible applications thereof.

The model has a dual purpose in that it serves as a further tool to compare the CGA process to the amalgamation process on a theoretical basis, whereby actual values for the free energy were calculated. Preliminary estimations have shown that the value of \( F_{II-I} \) is predominantly influenced by the value of \( \gamma_{23} \), which is the interfacial tension between oil and water and/or mercury and water for each process respectively. Actual measurements of \( \theta \) and \( \gamma_{23} \) were, however, done by calculations, at constant values of \( k \), it was found that the CGA process yielded lower values for the free energy, and hence is the more stable of the two processes. It is therefore seen that the CGA process is a viable alternative to the amalgamation process in terms of thermal stability.

**Experimental**

An experimental programme was commenced by determining the standard conditions for effective operation, followed by a sensitivity analysis on selected parameters and hence, lead to a data base providing sufficient information about the process to determine the conditions for optimal performance. The optimal conditions were used as the basis for further experimentation, including studies such as recycling the coal-oil and/or the ore phase and performing CGA process on an industrial ore. Finally, a comparative study was done, whereby the CGA process was compared to the mercury amalgamation process.

**Experimental materials**

A synthetic gold ore of grade 7g/t, was prepared from a silica sample \( (d_{40} = 106 \mu m, \) ranging from 25-300 \( \mu m \)), and a fine gold powder of particle size 44 \( \mu m \) was used for this purpose. The coal used had a density of 834 kg/m³, consisted of 40% ash and was milled down to a particle size of 90 \( \mu m \). The coal-oil agglomerates were prepared in a separate step, whereby the oil, i.e. ethane oleate, was added to the suspension of coal particles and agitated for a few seconds. A collector, i.e. potassium amyl xanthate (PAX) was added in a concentration of 1g/100 ml and its purpose was to increase the oleophilicity of the gold particles and/or enhance the floatability of the coal-oil agglomerates so as to enable effective separation. The batch experiments were conducted in 150 ml bottles, which kept the samples at manageable sizes and facilitated the analysis thereof.
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**Standard experimental conditions**

The experimental conditions were compiled from the results of a series of trial and error tests to ascertain the exact quantities of the raw materials needed to successfully perform the Coal Gold Agglomeration process on laboratory scale. For the purpose of this study, the selected conditions are referred to as the standard experimental conditions for the CGA process, which was the platform from which the sensitivity analysis was performed and is defined below.

The amounts of coal, oil, ore and water needed were determined and the following ratios were used: coal:oil—100:20 (by volume), coal:ore—1:1 (by mass) and the ore slurry was prepared by utilizing 130 ml of distilled water for every 4 g of ore. The coal-oil phase and slurry was contacted for a 50 minute period using a laboratory scale mechanical shaker. The agglomerates were separated from the slurry by means of scraping it from the surface of the aqueous medium. Finally, the ore and agglomerate samples were dried, ashed and dissolved into aqua regia, and after further treatment analysed on a Graphite Tube Atomiser (GTA). A set of experiments were conducted under the above-mentioned conditions and an average gold recovery of 38.4% was obtained.

**Sensitivity analysis**

It was needed to perform a sensitivity analysis on the CGA process in order to determine the conditions needed for optimal performance. The following parameters were selected:

- the coal-oil phase and slurry were contacted for different periods of time
- the mode of contacting these phases, i.e. shaking, stirring and the rolling bottle technique
- the means of separating the coal-oil agglomerates from the slurry, i.e. scraping, sieving
- the amounts of coal, oil and water were altered within the ratios: coal:ore, coal:oil and water:ore
- and volume and concentration of PAX was changed to determine the significance of this additive

The conditions for optimal performance were found by lumping all the factors which caused gold recovery percentages to increase. All further tests were done under the optimized conditions, which will be discussed under the next section heading.

**Recycling**

The optimized CGA process was conducted as normal, after which two consecutive recycling studies were done, the first one being the recycling of the ore phase and the second one being the recycling of the coal-oil agglomerates.

**CGA on an industrial ore**

The CGA process was performed on a real ore to determine the effect of mineralogy of the ore on the effectiveness of the process. The experimental study started by performing the process on an industrial ore sample, received from the Western Area Goldmine in South Africa, and recycling the coal-oil phase, after which coal-oil and ore samples were analysed for gold. A further analysis was then done whereby both the untreated and pre-treated coal-oil and ore samples were subjected to X-ray Diffraction analysis to determine which minerals were present in each, before and after contacting the agglomerates with the ore slurry.

**Comparative study**

A comparative study was conducted, whereby the CGA process was compared to the Amalgamation process. Mercury amalgamation is a process utilized predominantly by small-scale miners, whereby mercury is used to amalgamate the free gold particles in the ore. The amalgam is roasted on an open fire to get rid of the mercury, which vaporizes at these high temperatures and the remaining reddish powder which mainly consists of gold and is then sold.

Laboratory scale batch tests were performed, whereby the amalgamation process was simulated under the same conditions used at the Centre of Mineral Technology (CETEM) in Brazil and these were as follows: 30% solids by weight; mercury/ore ratio: 1/20; Sodium hydroxide (NaOH)/ore ratio: 1/1000 and conditioning for two hours in a horizontal cylinder at 20 rpm. The purpose of the sodium hydroxide was to optimize the surface properties of the mercury with respect to the feed charge in the closed vessel. The ore used was a synthetic gold ore (7g/t) and gold powder (44 µm).

After performing the amalgamation tests, the amalgam was separated from the slurry by means of a separation funnel and both ore and amalgam samples were treated with aqua regia to dissolve the gold present in each. The resulting solutions were analysed on the Induction Coupled Plasma (ICP), an instrument which is able to analyse for gold in the presence of other metals, such as mercury, which is dissolved by aqua regia as well.

**Results and discussions**

**Sensitivity analysis**

Table I depicts a summary of the results obtained after performing the sensitivity analysis. Note that they are arranged in order of increased gold recovery.

It can be seen that factors such as an increased amount of oil and contact by the rolling bottle technique negatively influenced the CGA process in terms of the percentage gold recoveries. The increased amount of oil gave rise to the formation of unstable agglomerates which could not be separated easily and hence, the recovery process was not

<table>
<thead>
<tr>
<th>Conditions</th>
<th>% Gold recoveries</th>
</tr>
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<tbody>
<tr>
<td>Standard</td>
<td>38.4</td>
</tr>
<tr>
<td>Coal:oil (Oil amount was doubled)</td>
<td>12.07</td>
</tr>
<tr>
<td>Rolling bottle technique</td>
<td>37.00</td>
</tr>
<tr>
<td>Concentration of PAX (doubled)</td>
<td>41.02</td>
</tr>
<tr>
<td>Volume of PAX (doubled)</td>
<td>41.36</td>
</tr>
<tr>
<td>Coal:ore (1.5 times the amount of coal)</td>
<td>41.95</td>
</tr>
<tr>
<td>Water:ore (water amount was halved)</td>
<td>43.35</td>
</tr>
<tr>
<td>Stirring</td>
<td>49.45</td>
</tr>
</tbody>
</table>
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brought to its full potential. Furthermore, the rolling bottle and shaking techniques for contacting the coal-oil phase and agglomerates were not as effective as stirring the two phases, where stirring was carried out by a magnetic stirrer. Further factors such as the addition of PAX, an increased amount of agglomerates and a decreased amount of water had caused the percentage gold recoveries to increase. Focusing on the most efficient means of separation, it was found that scraping, as compared to sieving, was the more feasible method for effective separation. Finally, the contacting procedure was repeated for different periods of time, ranging from 10 to 50 minutes and it was clearly found that the percentage gold recoveries were at a maximum for contact times above 50 minutes.

Optimized CGA

As mentioned earlier, the conditions for most efficient performance of the CGA process was a compilation of the parameters that favoured maximal gold recoveries. A summary of these are as follows: a contact period of at least 50 minutes, contact by stirring, the addition of a collector and a decreased amount of water was used to prepare the ore slurry. The CGA process was repeated under the specified conditions and analysis was done by either the Graphite Tube Atomiser (GTA) or the Induction Coupled Plasma (ICP). These results are shown in Table II.

It is seen that overall gold recoveries as analysed by the GTA has increased from 38.4 to 44% (Refer to Table I). Although an improvement was observed, it was still a very low result and for this reason it was sought to utilize an alternative instrument, i.e. the ICP for analysis. All samples were treated in the exact same manner as before i.e by drying, ashing and dissolving them into aqua regia, the only difference being the instrumental analysis.

As seen from Table II, the percentage gold recoveries had increased to a promising 85% and it could be concluded that the low results in previous experiments were rather instrumental based than caused by human error. This conclusion was made from the fact that the total mass balance of the system as illustrated in Figure 1, could not be closed in the case of the GTA, whereas the total gold were accounted for in the ICP analyses. Needless to say, all further analyses were performed on the ICP. The results of all tests were fairly reproducible with an error margin of less than 5%.

Recycling

Recycling of the ore phase

The coal-oil agglomerates were contacted with an ore sample after which it was separated and the ore contacted with a ‘fresh’ coal-oil sample of the same quantity as before. Both the coal-oil agglomerate samples were analysed for gold. The first batch of agglomerates recovered an average of 37.9% of the gold from the ore phase and the second batch of agglomerates a further 4.25% of the gold that was originally in the ore sample before any contact. As seen from the low results, the analysis was done by the GTA and although these values are low, it could still be concluded from the results that the coal-oil agglomerates are capable of recovering gold from very low grade ores.

Recycling of the coal-oil phase

The CGA process was performed under optimized conditions and the coal/oil phase was separated as normal, after which these agglomerates were then contacted with a ‘fresh’ ore sample. It was the objective of this study to determine what the possibility was of increasing the gold loading onto the coal-oil phase. The analyses were done on the ICP, whereby all the untreated and pre-treated ore and coal-oil samples were analysed. Table III, below, stipulates the results:

From the tabulated results it is seen that the agglomerates recovered an average of 83.03% of the gold after it was contacted with the ore the first time and a further average of 75.6% gold was recovered after recycling the same coal-oil phase. Therefore, it is found that the coal-oil agglomerates had the capacity of recovering an average of only 7.44% less gold after it had been recycled once. Furthermore, it was found that the gold loading onto the coal-oil agglomerates was increased from an average of ±6 to 12 g/ton, which confirms that the recycling procedure is an effective operation and hence, serves as an incentive to promote the CGA process in terms of the economical viability thereof.

CGA on an industrial ore

As the CGA process was repeated on a real ore sample whereby the coal-oil phase was recycled afterwards, a XRD analysis was done on the untreated and pre-treated coal and ore samples to determine if minerals other than gold were recovered by the coal-oil phase. The results of the XRD analysis are depicted in Table IV.

As seen from Table IV, no gold was detected in any of the samples. The X-ray Diffraction analysis is a surface based analytic method, which does not quantify the minerals on a surface, but rather qualifies those present. Therefore, it is clear that no gold particles were detected on the surface of any of the samples, which is no proof that they do not exist in the matrix of the sample. However, it is seen that the coal-oil agglomerates picked up some other minerals, besides gold and these results merely state that the presence of other minerals could have an effect on the performance of the Coal Gold Agglomeration process. Through ICP analysis it was

<table>
<thead>
<tr>
<th>Table II</th>
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<tbody>
<tr>
<td><strong>The gold recoveries found from the CGA process as performed under optimal conditions</strong></td>
</tr>
<tr>
<td>Method of analysis</td>
</tr>
<tr>
<td>Graphite Tube Atomiser (GTA)</td>
</tr>
<tr>
<td>Induction Coupled Plasma (ICP)</td>
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<tr>
<th>Table III</th>
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<tbody>
<tr>
<td><strong>Results obtained after recycling the coal-oil agglomerates</strong></td>
</tr>
<tr>
<td>% Au recovered in coal-oil phase after first contact</td>
</tr>
<tr>
<td>83.03</td>
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<table>
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<tr>
<th>Table IV</th>
<th>The results on XRD analysis</th>
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<tbody>
<tr>
<td>ORE— before CGA</td>
<td>COAL— before CGA</td>
</tr>
<tr>
<td>Quartz</td>
<td>Pyrophyllite</td>
</tr>
<tr>
<td>Non graphitized carbon kaolinite traces of mica</td>
<td>Less pyrophyllite and pyrite and chlorite</td>
</tr>
</tbody>
</table>

possible to determine what the effect of the different minerals was on the recoveries by the CGA process. The results are tabulated in Table V.

Refer to Tables III and V for a comparison of the results. By carefully studying and comparing the results, it is seen that less gold is recovered into the coal-oil phase after performing the CGA process on a real ore sample (a decrease from 83.03 to 76.4%). Furthermore, less gold is recovered into the coal-oil phase after recycling with a real ore sample than with the synthetic ore sample (a decrease from 75.6 to 43%). It could then be concluded that the CGA process efficiency is inhibited when performed on a real ore sample, where the gold particles are in competition with minerals such as: quartz, pyrophyllite, pyrite and chlorite (Table IV) to attach themselves to the coal-oil agglomerates.

Subsequent tests were performed on a gravity concentrate with a head grade of 12% gold. Total recoveries, close to those obtained with a synthetic ore, were achieved.

**Comparative study**

It is not only the purpose of this study to investigate the CGA process, but also to assess the potential of this new technology as an potential alternative to existing small-scale mining operations, such as mercury amalgamation. Environmental legislation has become more stringent and techniques such as amalgamation have been researched in the finest detail so as to come up with ways to improve the environmental safety of this ancient technique. The gold mining operations in the underdeveloped areas, such as the Amazon region and North Africa depend on the amalgamation technique for recovering the gold from alluvial ores, where the miners are not highly skilled or educated which makes such a simple operation a very attractive means of income. Acute mercury poisoning is not only confined to the immediate surroundings of operation, but affects the lives of many people downstream, as fish eat the mercury and it is digested to methyl-mercury, which is even more dangerous than the vapours produced by roasting. Although it is not within the scope of this paper to discuss the details of the dangers of mercury, it was however necessary to provide the reader with some background as to why a valid technical and economical alternative to mercury amalgamation is researched.

After performing a batch of simulated mercury amalgamation experiments, the results were found to be very promising towards the CGA process, as seen in Table VI.

The analysis was performed by the ICP and the results are the average of 3–5 batch experiments. As seen from the results, the agglomerates recovered an average of 15% more gold than by mercury. Although the CGA process does not outperform the amalgamation process in *sensu stricto*, it definitely is the better alternative based on the environmental friendliness and safety thereof. If referred to the THEORY section of this paper, it is seen that the CGA process is not only the better in terms of recoveries, but a free energy analysis has shown it to be the more stable process in theoretical terms.

**Conclusions**

The CGA process yielded promising results, which under optimal conditions was up to 85% gold recoveries. By performing a sensitivity analysis, the optimal conditions were found to be: contact periods above 50 minutes, contact by stirring, the addition of a collector and a decreased amount of water. Furthermore, factors such as an increased amount of oil and contact by the rolling bottle technique caused a decrease in gold recoveries.

A set of recycling experiments was performed, whereby the ore phase was recycled and it was found that the CGA process is suitable for recovering gold from very low grade ores. Further recycling of the coal-oil phase has proved that it is possible to increase the gold loading onto the coal-oil agglomerates, which will be to the advantage of the economical viability of the process.

The CGA process was repeated on a real ore sample, whereby the effect of other minerals was studied. The gold recoveries were less than with the synthetic ore and by performing XRD analysis on the coal-oil phase, it was found that the gold particles were in competition with minerals such as quartz, pyrophyllite etc. which attached themselves to the agglomerates, hence the lower results.

Finally, a comparative study was conducted, whereby the CGA process was compared to mercury amalgamation. Laboratory scale amalgamation tests were performed,

<table>
<thead>
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<th>Table V</th>
<th>The results obtained by performing the CGA process on a real ore sample and then recycling the coal-oil phase</th>
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<tbody>
<tr>
<td>First contact</td>
<td>Second contact</td>
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<tr>
<td>% Au recovery</td>
<td>76.4</td>
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<table>
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<tr>
<th>Table VI</th>
<th>Gold recovery percentages measured after performing Mercury Amalgamation compared to that of Coal Gold Agglomeration</th>
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<tbody>
<tr>
<td>Process type</td>
<td>% Au recovery</td>
</tr>
<tr>
<td>Mercury amalgamation</td>
<td>70</td>
</tr>
<tr>
<td>Coal Gold agglomeration</td>
<td>85</td>
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whereby gold recoveries of ±70% was found, which were less than for the CGA process.

Acknowledgement
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References