

Oxygen Mass Transfer in the Albion Process™: from the Laboratory to the Plant

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ABSTRACT

The successful commissioning and ramp up of the Albion Process™ at the GPM Gold Project relied on the successful scaling up of the process from batch and continuous pilot plant campaigns (Voigt, 2016). Critical information about reaction kinetics and residence time, grind size and pulp density were determined at the laboratory scale and successfully applied to the commercial scale. A limitation of small scale testwork, is that some parameters cannot be measured reliably and scaling up is a function of the physical size of the equipment which isn't possible to test with laboratory scale equipment. Oxygen mass transfer rate is one such parameter since this is a complex interaction of many factors including slurry temperature, solution and slurry chemistry, slurry viscosity, agitator type, dimensions and power, oxygen bubble residence time, oxygen purity, tank geometry and oxygen injection technique. Oxygen generation represents an important operating cost for the Albion Process™. Pivotal to the Albion Process™ operating economically at atmospheric pressure is the capability to efficiently transfer oxygen while utilising as much oxygen injected to the process as possible. To respond to this Glencore Technology developed the HyperSparge™ supersonic gas injector. This paper compares the HyperSparge™ against other sparging techniques to quantify the benefits of oxygen injection via a supersonic gas jet on scale up of the oxygen mass transfer system. The paper then examines plant survey data from the GPM Gold Project to demonstrate the very high oxygen utilisation that can be achieved with a correctly designed oxygen mass transfer system.

INTRODUCTION

One of the main challenges in chemical reactor design is the scale-up of processes from the laboratory to industrial scale. One particular challenge are those processes which cannot be faithfully simulated with an experiment at the laboratory due to the physical dimensions and complexity of the real system. One example of this is the design of oxygen mass transfer systems in atmospheric oxidative leaching such as the Albion ProcessTM. Some of the problems with the small size of equipment are the specific agitator power input is artificially high, the bubble residence time in the vessel is artificially low, the oxygen partial pressure at the base of the vessel is artificially low and the way oxygen is injected into the process may not be the same as the industrial process. Some of the problems of the complexity of the system is the dynamic nature of the process including the presence of solids, recycle streams and minor elements plus the variances within the reality of a process plant such as disruptions or variance of feed quality.

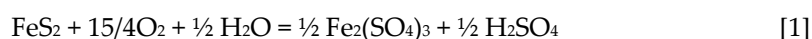
It is critical that for such processes the fundamentals are understood so that key data can be collected from appropriately designed experiments at the laboratory scale for input into a proven design approach to render the industrial scale process effective and fit-for-purpose.

To maximise oxygen mass transfer efficiency, operational availability, simplicity and safety, Glencore Technology (GT) developed the HyperSpargeTM supersonic gas injector. Originally for injecting oxygen to the Albion ProcessTM, the HyperSpargeTM has found application in other processes using air, oxygen, sulphur dioxide and gas mixtures. Over 400 HyperSpargeTM units are installed globally in duties from in-line slurry conditioning, fermentation, waste-water treatment, CIL/CIP processes and oxidative leaching.

This paper compares the HyperSpargeTM to other gas injection techniques such as ring spargers and converging nozzles. The key data are identified for scale-up from the laboratory to industrial scale. Survey data are examined from the GPM Gold plant in Armenia where the oxygen mass transfer system was successfully designed and comments are made about the operational advantages of the HyperSpargeTM compared to other systems such as sintered spargers.

OXYGEN MASS TRANSFER

The Albion ProcessTM is an atmospheric oxidative leaching process developed by GT in 1994 and is described extensively in the literature (Hourn & Turner, 2010; Hourn & Turner, 2012; Hourn et al., 2014; Voigt et al., 2015; Senshenko et al., 2016). Oxidation of pyrite is one of the key reactions in the Albion ProcessTM chemistry as shown in Equation 1.



The first step of the oxidation mechanism is the dissolution of oxygen in the liquid phase since electron transfer occurs in part through the action of the ferric and ferrous ion couple and the action of dissolved oxygen reacting directly with pyrite. Hence the rate at which oxygen is transferred is critical to the process efficiency and can be simplified for a reactor as shown in Equation 2 (Middleton & Smith, 2004).

$$\text{O}_2 \text{ Transfer Rate} = k_L a \cdot V \cdot (C_{\text{sat}} - C) \quad [2]$$

Where k_L is the liquid film transfer co-efficient (m.s^{-1}), a is the specific gas surface area (m^{-1}), V is reactor volume (m^3), C_{sat} is the oxygen solubility at saturation (g.m^{-3}) and C is the steady state oxygen level (g.m^{-3}). The terms k_L and a are normally combined to represent the oxygen mass transfer co-efficient which is the parameter to be maximised.

For practical purposes the k_La needs to be maximised by determination of a relationship with power input through the sparger and the agitator and then selection of equipment that satisfies the oxygen transfer rate requirements of the system. Middleton (1992) simplified the equation relating k_La to power input is shown in Equation 3.

$$k_La = K \cdot (U_s)^a \cdot (P_g/V)^b \quad [3]$$

Where K is a coalescence constant, U_s is superficial gas velocity (m.s^{-1}) and P_g is absorbed power (W). A correction is also applied for temperature since increasing temperature will reduce slurry viscosity and surface tension increasing the mass transfer interfacial area. The coefficients a and b are empirical constants that are system specific and are determined through testwork.

There are many theoretical correlations between k_La and energy input reported in the literature and the majority tend to be based on data in biochemical reactors meaning the extension to oxidative leaching can be problematic with limited applicability because the agitation intensity, system chemistry, operating temperature and oxygen injection methods are very different (Van't Reit, 1979; Vasconcelos et al., 1998; Oguz et al., 1987; Moo-Young & Blanch, 1981). Most of these correlations are also derived at very small scale, with laboratory vessels of 10 litres or less in size.

The best way to achieve successful industrial scale up is to determine the a and b empirical constants for equation 3 for a given system through specific experiments that reflect the actual system conditions. Once determined for a specific system, Equation 3 is then used along with other reactor design equations and experience to calculate the tank geometry and power input required from the agitator and gas injector to achieve the required k_La . GT has developed an experimental procedure to determine the a and b empirical constants for a given system which will result in a successful scale up with minimal error. An example of using this method to compare the experimental and calculated k_La for a copper leach system is shown in Figure 1.

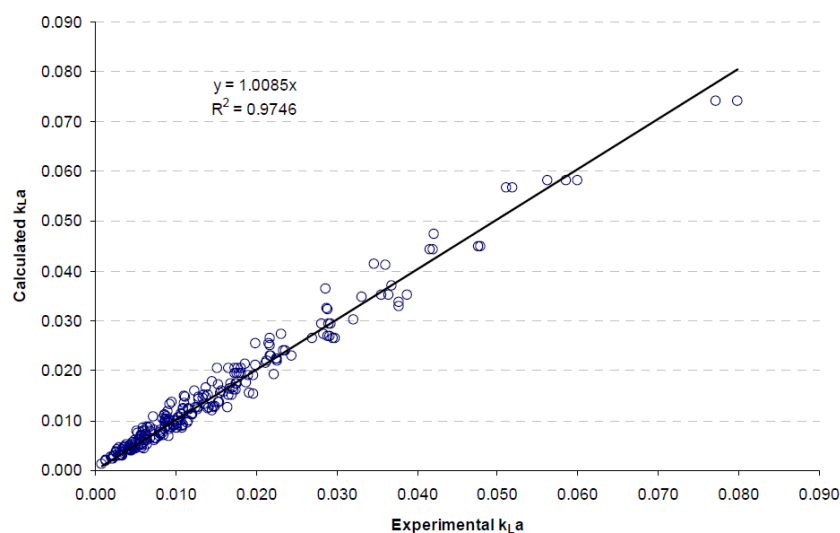


Figure 1 Experimental and calculated k_La for a system using GT experimental methodology

INJECTION OF OXYGEN

Importance of oxygen injection method

The method in which oxygen is injected has a large bearing on the efficiency of the oxygen mass transfer. Referring to Equation 2, the liquid film co-efficient (k_L) is proportional to the oxygen diffusion coefficient and inversely proportional to the liquid film thickness. The consequence for oxygen injector selection is that high pressure gas injection maximises shear and significantly erodes the liquid film thickness resulting in increasing k_L . Referring to Equation 3, the surface area (a) is proportional to power absorbed into the system relative to system volume and superficial gas velocity. Surface area is also inversely proportional to bubble size. The consequence for oxygen injector selection is to maximise shear and minimise bubble size but also to select an agitator that provides sufficient complimentary power input.

Options for oxygen injection

There are many gas injectors available on the market. The converging nozzle type and ring spargers (essentially open pipes) and among the most common from an operational perspective. Sintered spargers find application in flotation machines and chemical reaction systems and are well suited to the laboratory but are plagued with operational issues at the industrial scale, particularly in processes with chemical reactions due to the fast accumulation of reaction products and the continual need to clean them.

GT performed a number of experiments comparing the performance of an open pipe sparger, a converging nozzle type sparger and a newly developed supersonic sparger by GT, the HyperSparge™.

Open pipes and sparge rings do not transfer a significant amount of energy into the leach vessel, with all the energy required for oxygen mass transfer provided by the impeller. For these spargers, the gas is compressed to just above the hydrostatic head level in the slurry tank and the pressure drop across the tip of the injection point is low, resulting in low gas speeds.

Convergent nozzles, such as the CPT Slamjet and Minnovex gas spargers, were developed for flotation column aeration duties, and have found some use in slurry oxidation systems. These nozzles converge to a narrow opening, and the gas is compressed to several atmospheres above the hydrostatic head level in the slurry tank. The pressure drop across the tip of the injection point is 3 – 5 atmospheres, resulting in higher gas speeds at the point of injection. This results in more energy being put into the gas from the sparging system, and so less energy is required at the impeller.

Testwork carried out by GT has found that for oxygen mass transfer purposes, energy input to the system via the gas sparging system is more efficient than energy input via the impeller, as long as the majority of the energy released from the gas pressure drop across the sparger is converted to kinetic energy and not lost as heat. As such GT developed a supersonic gas injector, the HyperSparge™, to further improve the efficiency of the gas injection system. The HyperSparge™ used a converging-diverging nozzle to maximise energy recovery from the gas pressure drop by accelerating the gas to supersonic velocities.

Efficiency of oxygen injection methods

Energy is put into the gas sparging system by pressurising the gas prior to the sparger, to store energy in the gas stream. The theoretical energy input to compress the gas is defined in Equation 4.

$$W = (n/(n-1))mRT[(P_2/P_1)^{(n-1)/n} - 1] \quad [4]$$

Where n is isothermal efficiency, W is work (J), P_1 is outlet pressure (N.m^{-2}) and P_2 is inlet pressure (N.m^{-2}), T is temperature (K), R is the ideal gas constant ($8.314472 \text{ J.K}^{-1}.\text{mol}^{-1}$) and m is moles.

The pressure energy is then recovered as work into the system, ideally the efficiency of this conversion is maximised. High speed gas injection maximises turbulence at the injection point, which reduces the liquid film boundary layer around the gas bubble, increasing the liquid film diffusion rate, and so increasing the k_L value. The higher kinetic energy of the gas will translate to a higher interfacial area, which also assists mass transfer.

In a conventional pipe or ring sparger, the nozzle diameter is relatively large, resulting in a low velocity gas flow, which remains constant along the length of the nozzle. In this configuration, higher gas flows can lead to larger bubble sizes and consequently smaller interfacial areas (a). As a general rule, the size of the bubble generated from a conventional sparger will be close to the size of the orifice.

The formation of fine bubbles requires a specific type of nozzle to be fitted to the end of the sparger. Nozzles can normally be described as *convergent* (narrowing down from a wide diameter to a smaller diameter in the direction of the flow) or *divergent* (expanding from a smaller diameter to a larger one in the direction of the flow).

Convergent nozzles accelerate subsonic gases, with the dynamic pressure decreasing from the nozzle opening to the narrowest point as the gas is accelerated. Once the ratio of the gas pressure at the nozzle throat to the feed pressure reaches a critical value, the flow will reach sonic velocity. This forms a shock wave in the nozzle throat. The nozzle is then said to be choked, and the gas flow is at Mach 1. Gas flow cannot be accelerated beyond Mach 1 in the throat of the nozzle.

Increasing the nozzle feed pressure further will not increase the velocity of the gas at the throat of the nozzle, and so any additional pressure energy is then stored in the gas in the form of heat.

Divergent nozzles decelerate subsonic gases; the gas expands in the nozzle and slows down. However, when the gas enters the divergent section at Mach 1, the divergent nozzle will accelerate the gas, as the stored heat energy is converted to velocity. Combination convergent-divergent nozzles, such as those used in the HyperSparge™ can, therefore, accelerate gases that have choked in the convergent section to supersonic speeds. These nozzles develop thrust by converting the stored heat that builds up in the gas in the throat into velocity when in the divergent section of the nozzle.

When gas is accelerated to velocities above the speed of sound in the divergent section of the nozzle, the local pressure drops further, and does not recover to ambient pressure until the gas passes through the resulting shock wave as illustrated in Figure 2.

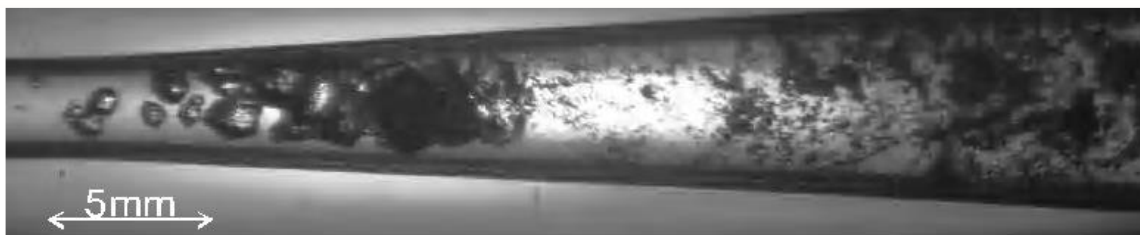


Figure 2 Collapsing bubbles crossing the shock wave of a supersonic nozzle (Kawamura et al., 2004)

The low pressure that is generated in the divergent section of the nozzle leads to collapse of the gas bubble into a fine mist, and results in a dramatic increase in the gas surface area. This is shown in the figure above, where the gas bubble leaves the throat of the nozzle at a size that is fairly similar to the size of the throat diameter, and then collapses to a fine mist of micro-bubbles in the low pressure zone in the divergent section. This collapse is triggered by the gas crossing the shock wave that is set up once the gas is choked in the throat of the nozzle. This shock wave can extend anywhere from the nozzle throat to the end of the nozzle and beyond, depending on the pressure applied to the nozzle.

A convergent-divergent nozzle has two important roles. The design of the nozzle determines the exit velocity for a given pressure and temperature. And because of flow choking in the throat of the nozzle, the nozzle design also sets the mass flow rate through the nozzle. Therefore, the nozzle design determines the thrust of the gas injection system. By changing the shape of the nozzle and the flow conditions upstream and downstream, you can control both the amount of gas that passes through the nozzle and the thrust generated by the nozzle. A convergent-divergent nozzle is represented below.

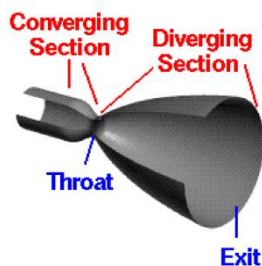


Figure 3 Cross section of a convergent-divergent nozzle (NASA, 2015)

EXPERIMENTAL

A number of experiments were performed to compare the nozzle types evaluating on the variables of thrust and oxygen mass transfer co-efficient (k_{La}). Experiments were performed in a jacketed, 1750 litre tank fitted with four baffles and a lid to minimise evaporative loss. The process solution employed was a copper leach solution at pH 1, with 15 g/l⁻¹ copper. High purity gaseous oxygen and nitrogen was used. The nozzles employed for the experiments were:

- 10mm open pipe
- Converging high pressure sparger, 4mm exit diameter

- Converging-diverging HyperSpargTM nozzle, Mach 3 (MV), 1mm throat and 5mm exit diameter
- Converging-diverging HyperSpargTM nozzle, Mach 1 (MT), 1mm throat and 1.7mm exit diameter

Thrust

Thrust is a direct measure of the efficiency of conversion of pressure energy in the incoming gas stream into momentum. The higher the thrust measured, the more efficient the nozzle will be in converting pressure energy into mass transfer without loss to heat dissipation. The nozzles were positioned perpendicular and centred to a balance surface. The gas plume impinged the balance's surface and the mass imparted recorded. For an identical energy input in the sparging gas (ie fixed flow and delivery pressure), each nozzle generated a different degree of thrust. The data are shown in Figure 4.

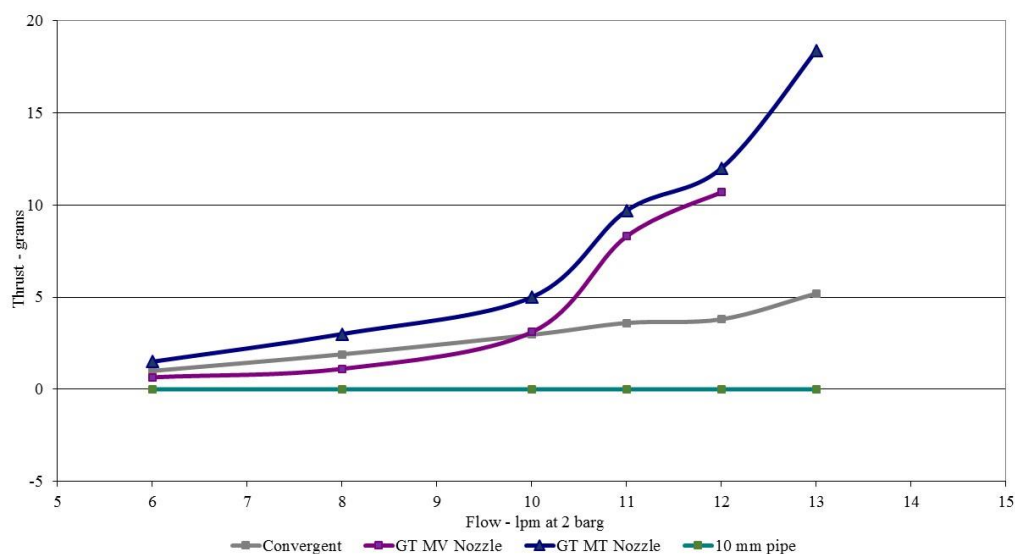


Figure 4 Developed thrust measured at 200kPa

The results above show that at low oxygen flow all of the profiled nozzles exhibited similar thrust values; less than 3 grams (g) of thrust recorded by the scale. However, at higher flows, the nozzles have a very different behaviour. The open pipe sparger did not develop any thrust at all across the entire range of gas flowrates, and so did not exert any force on the balance. The convergent nozzle showed an almost linear response, with thrust increasing with flowrate, up to a maximum of 5 g at 13 lpm of gas flow.

Both GT HyperSpargTM supersonic nozzles generated significantly more thrust than the open pipe or convergent nozzle. In general, the nozzle designed to produce maximum thrust outperformed the high velocity nozzle across all flow rates, achieving a thrust of 18 g at a gas flowrate of 13 lpm.

This compared well to the maximum theoretical thrust for a nozzle discharging 13 lpm of gas flow of 21 g. This meant that 85 % of the pressure energy in the gas stream was converted to momentum

for the HyperSpargTM MT nozzle profile. Both supersonic nozzles also displayed a considerable steepening of the thrust curve at gas flowrates above 11 lpm, close to the point where the nozzles were choked, indicating superior thrust as the gas speeds exiting the convergent-divergent nozzles approached supersonic speeds.

Oxygen mass transfer co-efficient (k_{La})

For temperatures up to 50°C oxygen mass transfer co-efficient was determined using the Dynamic Gassing Out Method (Taguchi and Humphrey, 1966). Above 50°C the combined sulphite method was used (Puskeiler, 2005). Experiments were conducted with and without solids in the reactor. The results are shown in Figure 5.

The GT MV and MT nozzle designs resulted in a consistently higher k_{La} value across the entire range of gas flow rates tested relative to the open pipe and the convergent nozzle. As the gas flow rate approached choked flow in the GT HyperSpargTM MV and MT nozzles, the difference in k_{La} was more pronounced, and was up to 300 % of the value observed for the conventional open pipe and 50% compared to a converged nozzle.

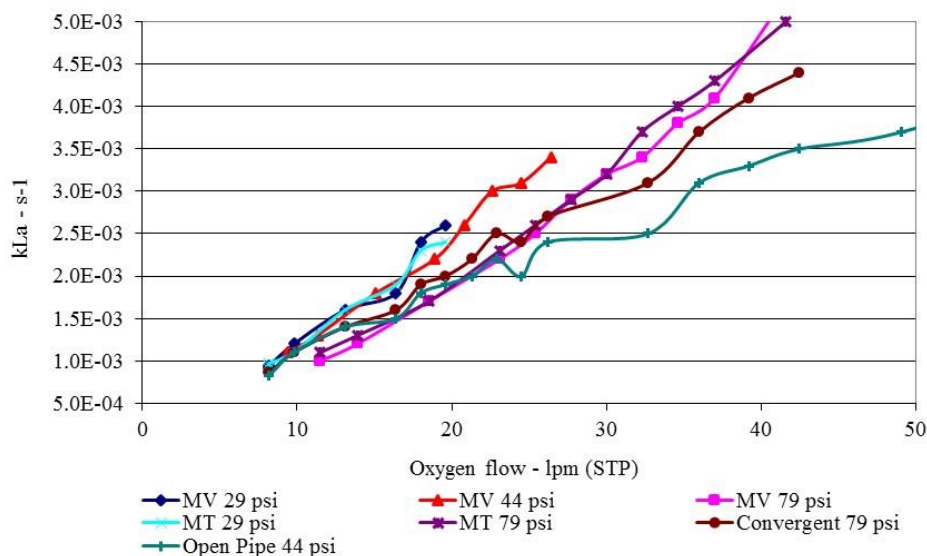


Figure 5 k_{La} at different supply pressure

The GT HyperSpargTM MV and MT nozzles showed similar behaviours because both are convergent-divergent type nozzles. Of the two HyperSpargTM nozzles, the MT nozzle provided the highest k_{La} , gas utilisation and thrust values, typically by 25 % relative to the MV nozzle at a set gas flow rate. The higher thrust generated in the MT nozzle at the expense of velocity explains this difference.

SCALE UP TO INDUSTRIAL SCALE

Through the scientific process, GT established that supersonic gas injection was technically a superior way to inject gas into a slurry requiring oxygen. The next step was to develop the converging-diverging nozzle into a product that can be used at the industrial scale.

The HyperSparge™

The HyperSparge™ was developed by GT as a complete gas injection solution. At the heart of the device is a converging-diverging nozzle fabricated from a specially selected wear-resistant ceramic which accelerates gases to supersonic velocities for injection into solutions and slurries. The nozzle is designed to minimise pressure energy loss. A schematic of the HyperSparge™ and close up of the nozzle is shown in Figure 6.

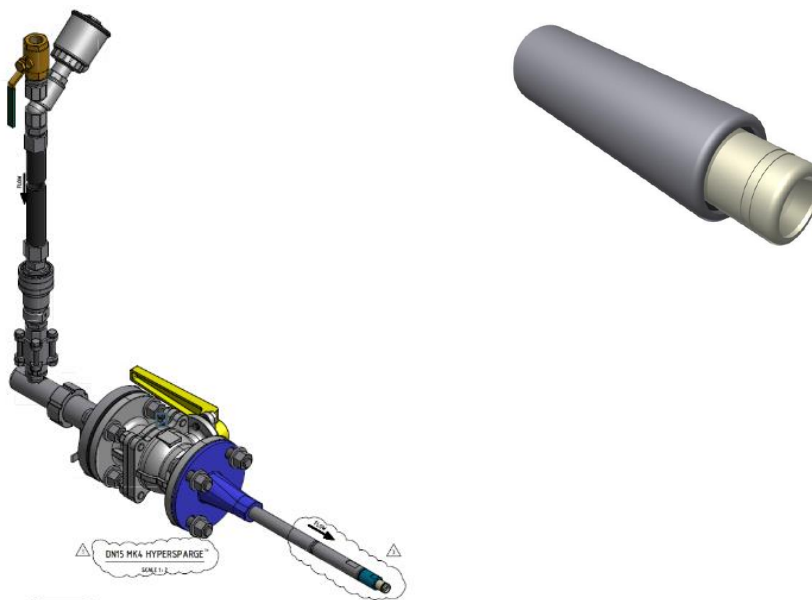


Figure 6 HyperSparge™ supersonic gas injector and nozzle

Apart from the advantages of superior oxygen mass transfer, the HyperSparge™ is safe and easy to use. The HyperSparge™ can operate over a range of pressures which can be adjusted to regulate flow. The nozzles can be easily interchanged with the threaded nozzle holder allowing the use of different nozzle diameters and hence flow operating envelopes. One attractive feature is that the sparger can be inserted and removed from a vessel while it is full of slurry spillage free without using any additional removal device plus with a throat diameter of typically 7 mm, the sparger does not become blocked and self-cleans by the action of the gas flow. This means less operational downtime and process interruption.

Oxygen Mass Transfer System Design

During the GPM Gold Albion Process™ plant development, key oxygen mass transfer experiments were conducted to determine the k_{La} under conditions as close as practicable for temperature and slurry conditions. The data was used by GT to then correctly size the gas injectors, agitators and select ideal tank dimensions. The design criteria for the GPM Gold Plant is shown in Table 1.

Table 1 GPM Gold Albion Process™ Plant Design Criteria

Parameter	Units	Nominal	Design
Throughput	tph	13.1	14.5
S ²⁻ concentration	%	17.6	20.0
S ²⁻ oxidation	%	76	76
Oxygen Utilisation	%	90	80
Required k_{La}	m.s ⁻¹	0.12	0.14
CIL Gold recovery	%	92	90

The economics of the industrial process rely on the efficiency of oxygen transfer to the system to maximise transfer rate (achieving design residence time) and maximise transfer efficiency (utilisation of the oxygen that is injected).

A survey was completed to collect plant data and assess the transfer rate and transfer efficiency. Since an industrial plant was the subject of the survey, the plant couldn't be taken off-line to replicate laboratory experiments. Instead, the oxidation extent and oxygen consumption from the operating plant were compared against the design. The conditions of the survey were a throughput rate of 14.0 tph concentrate and a sulphide concentration of 17.0%. The survey data for oxidation extent and gold recovery are shown in Figure 7.

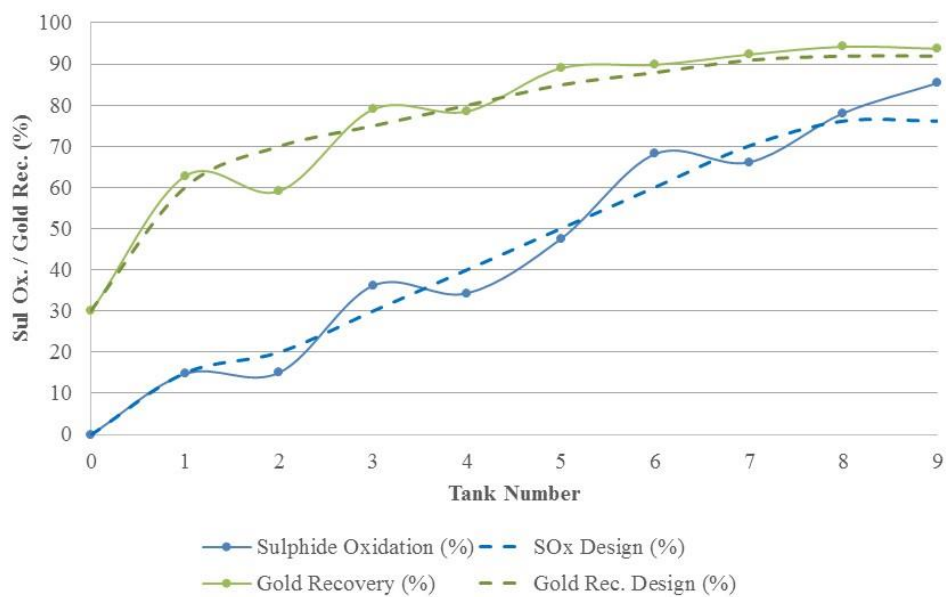


Figure 7 Sulphide Oxidation survey of GPM Gold Albion Process™ Plant.

Figure 7 shows that at the design throughput the sulphide oxidation is achieved along with corresponding gold recovery.

The oxygen utilisation was determined by comparing the oxygen injection to the process during the survey period against the modelled values. The results are shown in Figure 8.

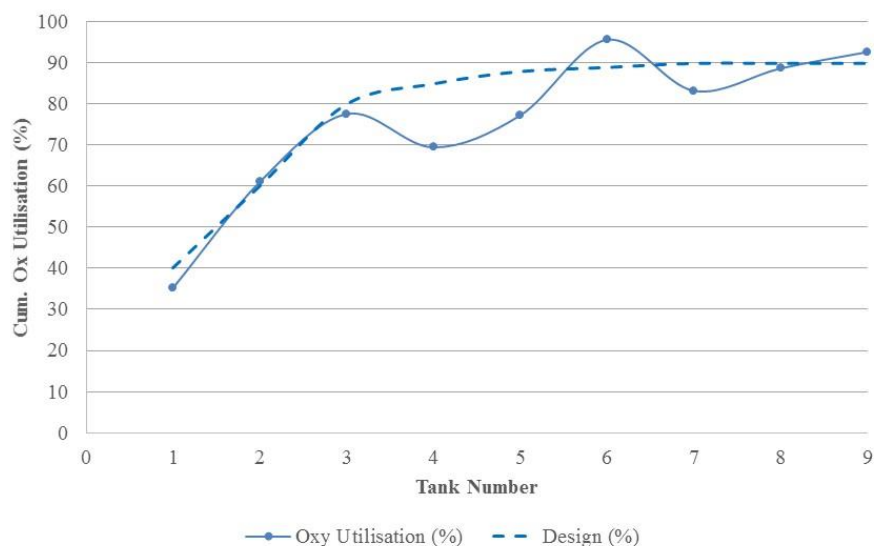


Figure 8 Oxygen utilisation at the GPM Gold Albion Process™ Plant

Figure 8 shows that the cumulative oxygen utilisation is in excess of the design value of 90% achieving 93%. This observation is consistent with other multi-tank Albion Process™ plants where it appears oxygen that is dissolved in the first few tanks of the leaching train is utilised later in the process.

Both Figures 7 and 8 illustrate that the oxygen mass transfer system was correctly and successfully designed.

CONCLUSIONS

Oxygen injection using convergent-divergent nozzles generate superior thrust and oxygen mass transfer compared to other gas injection techniques. Power delivered to the system is more efficient through gas injection rather than mechanical agitation. The HyperSparge™ is a development of the convergent-divergent nozzle and offers additional advantages over other sparging technologies contributing to a safer work environment, maximising process run-time and optimising energy input through the agitator. GT successfully scaled up the oxygen mass transfer system from the laboratory to the industrial scale at the GPM Gold Albion Process™ plant which achieves greater than design performance in terms of oxygen mass transfer and oxygen utilisation.

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