

THE APPLICABILITY OF CARBON CAPTURE AND SEQUESTRATION IN PRIMARY ALUMINIUM SMELTERS

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Abstract

Climate Change is affecting every industry including the Primary Aluminium industry. The International Energy Agency has developed several strategies how to abate GHG emissions on large industrial scale. One of the tools identified to abate CO₂ emissions from large sources is carbon capture and sequestration (CCS). Earlier papers have touched on the topic of using CCS in primary smelters and in this paper further details are shown of technological aspects of CCS, but also on some of the specific conditions that have to be dealt with when applied in primary aluminium smelters. Conclusions are presented.

Introduction

Today, Climate Change is generally accepted as a force behind a change in our Earth's atmosphere. This paper will look at a specific technical aspect of abatement of GHG emissions that is becoming a standard for many industries: carbon capture & sequestration (CCS).

While CCS is generally regarded as tool for industries like the Power Generation and Iron & Steel industries, a sector based approach [1] in new international protocols may require owners and engineers of primary aluminium smelters to investigate in more detail the technical applicability of CCS in primary smelters. A closer look at the summation of GHG emissions from primary aluminium shows that the main GHG emission is linked to the CO₂ emission from power generation. A smaller portion is related to direct CO₂ emissions from the use of carbon in the reduction process. While the latter is much smaller, it is the main CO₂ source that is directly within the span of control of the primary smelter. Hence, we apply CCS to these emissions.

There is an incredible amount of work in progress to demonstrate CCS technologies all over the world. Different technologies are tested while others are in early stage of development. In this paper we present some of the basics of CCS so that there is a better general understanding on how this applies in primary aluminium smelters should it be introduced at some point in the future.

GHG Emissions from Primary Aluminium Smelters

There are various publications available that describe the GHG emissions from primary aluminium smelting. In general, the emissions are reported in the CO₂ equivalent as if each emission is CO₂. Because within the numbers, different people use different conversion factors, such as for the GHG intensity for electric power, there are some differences within the various published numbers. In this paper we refer to the data presented in the 2009 *Light Metals* paper by Lorentsen et al [2].

The best method of presenting GHG emissions of a particular production process is to use the life-cycle methodology. This way all main aspects of the process are included even if they are indirect, such as generation of power in a power station.

If we review the direct emissions in a primary smelter then we have to consider those emissions that are generated within the fences of that smelter. The data presented is a reference case of a modern 350,000 tonne per annum smelter utilizing point feeder prebake anode (PFPA) type cells nominally operating at 340 kA. Anodes are prepared on-site and baked in a modern open-type furnace. The baking furnace has a modern dry alumina scrubbing type of emissions control system. Firing of a fuel oil or natural gas results in a flue gas flow in order of 80,000 Nm³/h with 8 vol% CO₂. Over time the flue gas ducts may degrade and ingress of air dilutes the combustion gases. The volume flow may increase to about 250,000 Nm³/h. Relative to air, the combustion gas is the main source of CO₂ and thus only a dilution takes place.

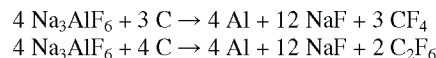
Table I. Life-cycle GHG emissions from a modern primary aluminium smelter (Source: Lorentsen et al^[2])

Source	Hydro Electric Power	Gas-fired Power	Coal-fired Power
	kg CO ₂ eq/kg Al	kg CO ₂ eq/kg Al	kg CO ₂ eq/kg Al
Alumina production	1.80	1.80	1.80
Anode production	0.30	0.30	0.30
Electrolysis - Carbon	1.50	1.50	1.50
Electrolysis - AE	0.30	0.30	0.30
Casthouse	0.06	0.06	0.06
Electric power	0.00	5.80	13.60
Sum	3.96	9.76	17.56

In the electrolysis process, two types of GHG emissions are identified. The first is the consumption of the carbon from the anodes. Based on a net carbon consumption of 410 kg carbon per tonne of metal, the contribution is 1.5 kg CO₂eq per kg of metal.

The second is the formation of perfluorocarbons or PFCs. These are formed during the anode effects that occur during reduction. Local imbalances in the molten cryolite create conditions that lead to some side reactions of which the gas CF₄ and C₂F₆ are the main products. Relatively, these are generated in small quantities but because their Global Warming Potential is large, 6500 and 9000 CO₂ equivalents respectively, they cannot be ignored.

Equation 1: Fundamental reactions of anode effects



It is found that the generation of PFC is directly linked to the frequency and duration of anode effects. In practice, the best method to minimize these effects is to avoid the imbalances by applying good process controls. New smelters have these controls and have demonstrated that they can operate with PFC emissions that are in order of 0.3 kg CO₂eq per tonne Al.

In the cast house a small amount of fuel or electricity is used in the various furnaces to handle the molten metal before casting. This contributes to about 0.06 kg CO₂eq per kg Al.

CO₂ Balance for the Reference Case

For our reference case primary smelter we can draw up the following CO₂ balance.

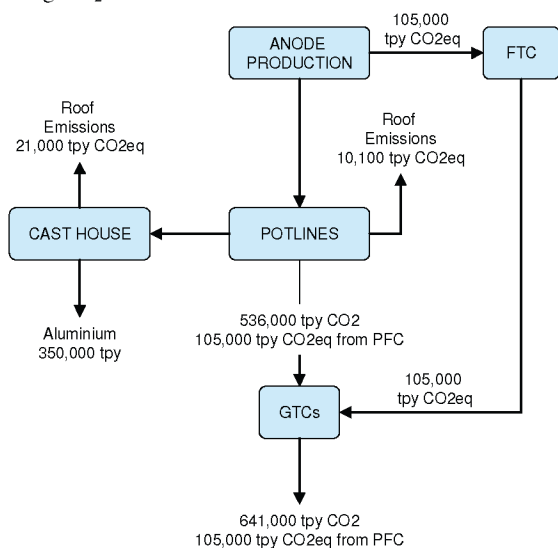


Figure 1. CO₂(eq.) balance in a modern primary smelter

Some CO₂ and PFC escapes through the hooding of cells when opened or through openings. In the reference case, it is assumed a hooding efficiency of 98 percent, which results in the roof emissions of 10,100 tpy CO₂eq.

In Figure 1 can be seen that in this model the fumes from the FTC is taken to the GTCs and blended in. The CO₂ amount in these fumes represents more than 10 percent of the total and should not be ignored for this investigation into the application of CCS.

Carbon Capture Technology

Carbon capture technology based on amine-type solvent has been around for many years. It is one of the standard gas cleaning technologies in the oil & gas industry. What is different is that for power plants and for aluminium smelters the gas to be treated is much larger in flow, but, moreover, contains oxygen (O₂). Oxygen is detrimental to conventional solvents used to absorb the CO₂. Only modified solvents can be used. A further difference is that in aluminium smelters the CO₂ concentration is relatively low at levels around 1 vol%.

At present, there are two key technologies considered for removal of CO₂ from fumes containing oxygen. The more conventional process is the use of modified amines and the more emerging

process is the so-called Chilled Ammonia process. Both of these will be discussed further.

CO₂ absorption by means of amine-type solvents

Amines have been used the most in the conventional oil & gas industry. It is a solvent dissolved in water (30 wt%) in which CO₂ absorbs and reacts with the solvent. In anticipation for the use in oxygen containing flue gas, the chemical companies such as DOW and BASF have developed modified amines and amino acids to control the effective capacity to absorb CO₂. Amines degrade in so-called heat stable salts which are not taking part in the CO₂ absorption process. Hence, it becomes an impurity that needs to be removed from the process. It also means that fresh solvent needs to be added continuously, which adds significantly to the operational costs.

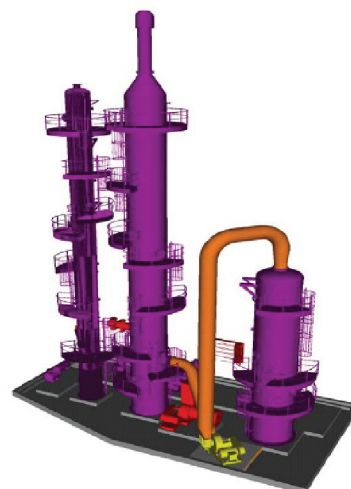
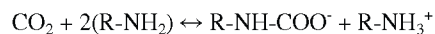


Figure 2. Application of the BASF PuraTreat solvent in a carbon capture unit at the refinery of Queensland Nickel to remove CO₂ from flue gas of a gas-fired boiler (Source: Hatch Ltd).

Typically, the fumes are pre-cleaned to remove acid gases such as SO₂ and HF that otherwise compete with CO₂ in the absorption process. Then the fumes enter the absorber where it passes through a number of layers with packing to enhance the contact between the rising fumes and the falling flow of solvent. During the contact the CO₂ is absorbed into the solvent. The absorption is most effective at lower temperatures; however, during the absorption heat is released from the physical process. This heats the solvent (and fumes) and in some cases external heat exchangers are used to provide for cooling within the stages.

Equation 2: Fundamental reaction in the amine process



Next, the CO₂-rich solvent is taken from the absorber to a regenerator for stripping of the CO₂. This done by heating the solvent to a temperature that does not exceed 130 °C in order to avoid excessive degradation. If this is done properly the CO₂ releases quick and an almost pure gas (some water vapor is left) can be taken from the stripper to a condenser to remove the water and produce a dry CO₂ product. The CO₂-lean solvent is taken from the stripper bottom back to the absorber. Heat exchangers are used to manage the energy balance and optimize heat input in the overall process. A special filter is used to extract the heat stable salts that are to be disposed of.

Key aspects to consider are:

- Solvent cost at \$800-\$1200 per tonne.
- The level of pre-cleaning to avoid other acid gases to interfere in the amine process.
- The amount of heat released in the absorption process.
- The height of the packing and associated resistance to gas flow through the absorber column.
- A source of heat to strip the CO₂. Typically, 2 to 4 GJ of energy is required per tonne of CO₂.
- Proper operation of the steam reboiler to avoid excessive degradation of the solvent (The main byproduct is bicine, which is known to cause plugging of equipment).
- Good heat recovery systems to manage the energy balance in the overall process.
- Deliver a dry CO₂ gas that is not corrosive.

CO₂ absorption by means of the Chilled Ammonia process

Much attention is given to the Chilled Ammonia process that is under development with Alstom Power. This process, an invention of Dr. Eli Gal who is responsible for many innovations in the field of wet flue gas desulphurization technology, aims to be as effective in removal of CO₂ but at much lower cost to build and to operate. Early calculations show a considerable energy saving when compared with the use of more conventional amine solvents. That this technology is most promising is attested by the unusual large number of pilot and demonstration plants that are either in design or early operational phase.

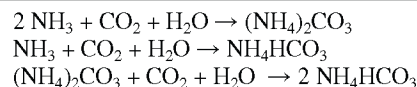


Figure 3. Chilled Ammonia demonstration plant at AEP's Mountaineer power station (Source: AEP).

The gases with the CO₂ first are deeply cleaned and chilled to a temperature below 10 °C before entering the absorber vessel. In this vessel the fumes are in contact with a slurry of water, ammonium carbonate and ammonium bicarbonate. To ensure the absorption of CO₂ takes place the pH is controlled greater than 7. The absorption of CO₂ is relatively slow and a packing is used to improve the contact between gas and slurry and it achieves over 90 percent of CO₂ removal.

The low temperature is required to keep any free ammonia in the slurry as its partial pressure is very low at that point. Normally, only a few ppm of ammonia is carried over with the clean fumes. For that, the clean fumes are washed with water to minimize the escape of free ammonia gas and to keep it within allowable limits.

Equation 3: Fundamental reactions in the Chilled Ammonia process



Under the process conditions in the absorber the ammonium forms a solid carbonate/bicarbonate mixture. For this, the absorber design is based upon the proven designs of FGD absorber towers. Similar to FGD operation, a bleed flow is taken to a set of hydrocyclones where the slurry is split into a solid rich underflow and a solids-lean overflow. The overflow is returned to the absorber.

The solids-rich underflow is brought up to high pressure and taken through a heat exchanger to heat the slurry to about 80 °C. By doing so all the solids go into solution so that clear liquor is obtained that is then taken to the regenerator.

In the regenerator a pressure is maintained in excess of 20 bar. By adding some heat the process conditions make the CO₂ liberate from the liquor. Under these conditions the phase diagram is such that an almost pure CO₂ gas is released. Only a small fraction of water vapor is carried over (2.5% vol) and all ammonia remains in the liquor. The CO₂ is separated and made available to the compression unit. The ammonia and water are recovered and re-used in the absorption process.

Key aspects to consider are:

- Solvent cost at \$100-\$200 per tonne.
- The level of pre-cleaning to avoid other acid gases to interfere in the chilled ammonia process.
- Chilling of the incoming fumes to temperatures around 10 °C.
- The amount of heat released in the absorption process, which is less than the amine process.
- The height of the packing and associated resistance to gas flow through the absorber column.
- A source of heat to strip the CO₂. The required amount of heat is around 1 GJ per tonne CO₂ produced.
- Good heat recovery systems to manage the energy balance in the overall process.
- Operating the regenerator at high pressure which is advantageous when a compression unit is included.
- Deliver a CO₂ gas that is not corrosive.

CO₂ Compression for Transport

In a CCS unit the CO₂ must have an ultimate destination. To get to its destination the CO₂ is compressed into a dense phase that makes it fairly easy to transport in a pipeline. There are a number of examples of CO₂ pipelines of which some are over hundreds of kilometers long.

For sequestration or enhanced oil recovery (EOR) applications the typical pressure at the discharge is 190 bar(g). For the transport of CO₂ from the Dakota Gasification Company (DGC) in Beulah

ND this requires a motor of 13 MW to send the CO₂ through a 320 km long carbon steel pipeline of 14 inches in diameter [3]. Some of the key challenges are related to mechanical integrity and balancing, materials of construction for seals, control system, inter-stage cooling, and contaminants in gas stream.

CO₂ sequestration and EOR require high pressure ratios up to 1:190 as in the case of DGC. This is achieved by multistage compression with interstage cooling. Step by step the CO₂ is elevated in pressure while kept in the right region of its phase diagram. This is explained in Figure 5.

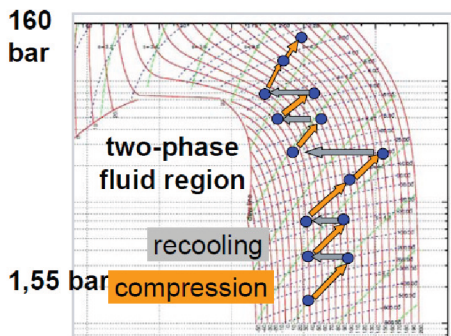


Figure 4: Performance diagram showing the different stages in compression of CO₂ [3].

Compression from 1 bar to 190 bar requires a power input of 13 MW for 125 tonne/hr of CO₂. This is a very large amount of energy. The Chilled Ammonia process delivers the CO₂ gas at a pressure of 20 bar which reduces the pressure ratio from 1:190 to 1:9.5 which is a considerable difference that results in a large energy saving.



Figure 5: 125 tonne/hr RG080/8 MAN Turbo compressor at the Dakota Gasification Company providing CO₂ to the enhanced oil recovery fields at Weyburn and Midale in Canada [4].

A development to follow is the introduction of a different way of compressing the CO₂ gas using a so-called RAMGEN Supersonic Shock Wave CO₂ Compressor [5]. This novel technology is derived from the aerodynamics that takes place in an aircraft engine such as the F-15. The physics are such that the required pressure ratios can be achieved in very few stages when compared to a conventional compressor system. One such unit in development delivers a pressure ratio of 1:100 in only 2 stages.

Geological Sequestration of CO₂

Although CO₂ is used in our daily lives in many ways and products, the quantities emitted from industry facilities are orders of magnitude larger. Sequestration of CO₂ in geological formations is one solution that helps us mitigate GHG emissions.

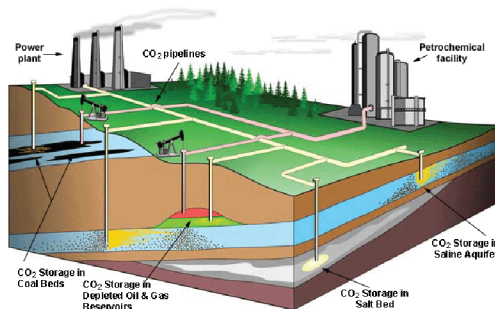


Figure 6: Options for geological sequestration of CO₂.

The following options are identified for sequestration of CO₂ (See also Figure 6):

- Coal beds incl. coal bed methane recovery
- Depleted oil and gas reservoirs
- Salt beds
- Saline aquifers

CO₂ used in Enhanced Oil Recovery (EOR)

An important question in a CCS application is what the end-use is for the CO₂. Very quickly one will find that only in a very few cases there is a business case to capture CO₂ in large amounts. One application that has been found to be viable is the use of CO₂ for enhanced oil recovery, also called CO₂ floods.

EOR has been applied for many years and only in recent years it has been dubbed as “sequestration” as a result of the large amount of CO₂ that remains caught in the oil reservoir. For example, in EnCana’s Weyburn EOR field, the injection rate is 5000 tonne per day. Of that 1600 tonne per day is co-produced with the fluid (oil and water) which means effectively 3400 tonne per day is stored in the oil reservoir. For Weyburn this is equal to 1 million tonne per year of CO₂ sequestration, which makes it one of the largest in its kind. In the US, however, the use of CO₂ for miscible floods in oil fields is more like 450,000 tonne per year on average. This is important because it means that the amounts of CO₂ required in EOR can be compatible with the amount of CO₂ captured from an aluminium smelter.

In the Weyburn (Saskatchewan, Canada) field water and CO₂ are injected in a mature oil field that had a declining oil production (See Figure 6). The CO₂ has low viscosity and effectively dilutes the oil like a solvent with the result that the mixture of fluids flows much easier through the pore space in the reservoir. The water acts as a barrier that pushes the fluids towards the other side of the reservoir where the producer well is located. Through this well the fluids are extracted from the reservoir and collected in the receiving facilities. Here the CO₂ is separated from the produced fluids and conditioned before recycled back to the injection wells of the plant.

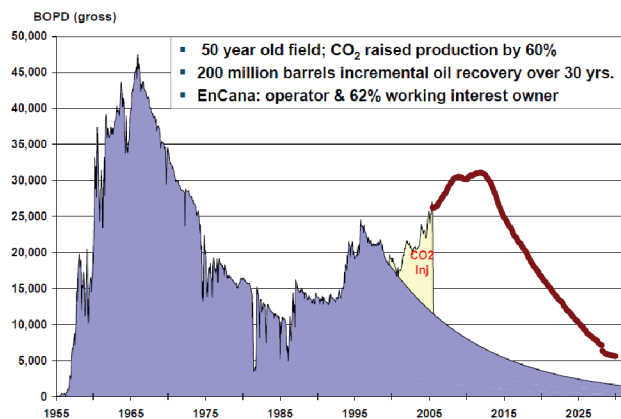


Figure 7: Impact of CO₂ injection to enhance oil production in EnCana's Weyburn field [6].

Figure 7 illustrates the impact of CO₂ injection in the Weyburn field. The incremental oil production provides for sufficient revenues to cover for the costs of using CO₂ while the net (to certain extent one has to account for the CO₂ released from the extra barrels of oil) CO₂ emissions are reduced.

Specifics of CCS in Primary Aluminium Smelters

Much is written about the application of CCS in fossil fuel fired power plants and not much of the applications in other industries, let alone production of Primary Aluminium. In this section we highlight some specifics that come with the application in primary aluminium smelters.

Composition of the fumes

The fumes are air drawn through the ventilation system. The starting CO₂ concentration is 385 ppmv or 0.0385 vol%. In the cells the carbon consumption is net 410 kg per tonne of aluminium. This equates to the formation of 61.2 tonne per hour of CO₂ that is picked up by the ventilation flow. The fumes are cleaned in the dry scrubbers and in the end the stack gases contain 0.85 vol% of CO₂. Compared with power generation units this is relatively low. Lorentsen et al [2] reported similar findings and proposed to seek smart solutions to increase the CO₂ concentration to comparable levels in power generation applications. This also has the potential to reduce the overall gas flows to save on capital costs due to the smaller size (diameter) of the gas handling equipment. However, in this paper we follow the more conservative path dealing with the full volume of fumes with "just" 0.85 vol% CO₂. It will remain the base case for any comparison in aluminium smelters.

A comparison of gas constituents is presented in Table 2 to show some of the subtle differences for application in aluminium smelters.

Table 2: Comparison of Gas Constituents (Typical values)

	Aluminium smelter (after GTC)	Coal fired power plant (after FGD)	Unit
CO ₂	0.85	14	vol%
H ₂ O	3	12	vol%
HF	0.5	1	mg/Nm ³
SO ₂	300	250	mg/Nm ³
SO ₃	<1	20	mg/Nm ³
Particulates	5	50	mg/Nm ³

In general the gas from the GTC is as clean as the gas from the FGD unit in the power plant. It is assumed that the GTC has good filters to keep the particulate low.

At first glance, it is not obvious that a certain advantage exists in aluminium smelters. The gases have always very low water vapor content. In a wet cleaning system this results in a low adiabatic cooling temperature of around 30 °C, which is 25 to 35 degrees lower than in a power plant. It may not seem large but this potentially saves considerable energy for cooling the gases down to target temperatures for Chilled Ammonia of 10 °C. Moreover, for amine systems it is likely that further cooling is not required.

Guidelines indicate that SO₂ levels need to be as low as 10 ppmv or 30 mg/Nm³ for application of CO₂ capture for both amines and chilled ammonia. This means that further cleaning is required. In an aluminium smelter this can be achieved by using technology that is already used at full scale such as in the smelter of Alcoa in Massena East, NY. Two wet scrubbers are known to have achieved SO₂ concentrations as low as 4 ppmv using soda ash as reagent.

One general assumption is made that, on the basis of very low solubility in water, gaseous CF₄ and C₂F₆ are inert and pass through all cleaning stages. With trials this needs to be verified. Also, new technology may be incorporated to remove these constituents as well.

Steam requirements

Normally, both CO₂ capture with amines or chilled ammonia require steam to provide the energy in the stripper/regenerator. This is a challenge because in aluminium smelters typically there is no steam available and certainly not in the amounts needed for the CO₂ capture units. This means that separate steam generators are required or that heat is found in another way by extracting this from the aluminium process. A quick calculation shows that this is in order of 55 to 75 MW of heat depending on the specific energy requirement for CO₂ separation in the stripper or regenerator. For the Chilled Ammonia process this is in order of 15 MW only.

CCS Integration in a Primary Aluminium Smelter

First, we need to consider a typical layout of a modern prebake smelter. For 350,000 tpy of metal there are two parallel buildings each holding two potrooms. Each line has a GTC unit for ventilation of the cells. A baking furnace is nearby and the FTC is not too far from the two GTC units.

One GTC treats 2,000,000 Nm³/h of fumes. When CCS is applied there is a choice of installing long ducts to a central CO₂ capture system or has individual CO₂ capture units placed adjacent to each GTC. Taking in consideration that pushing a total of 4,000,000 Nm³/h of fumes through a single CO₂ absorber is not practical and that two individual towers lead to absorber sizes that are consistent with modern FGD absorbers, the option of individual CO₂ capture units is preferred. However, in between absorber and stripper/regenerator distance is less critical so that a central CO₂ stripper/regeneration unit can be considered. This also results in a single compression unit to handle all CO₂ gases.

In the previous section it was made clear that also the fumes from the GTCs need to be deep cleaned before they can be allowed in

the CO₂ absorber. A basic approach is to install a dedicated wet scrubber down stream the GTC, again, just like in Alcoa Massena East is the case but then without the spare absorber. However, the effective cleaning inside such absorber takes limited space and using modern technology it may be possible to integrate this in the CO₂ absorber to make the system more compact and cost less.

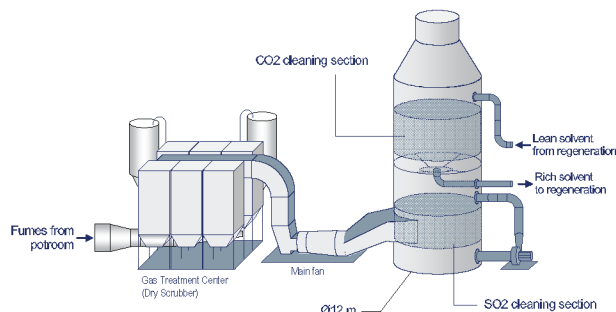


Figure 8: Integrated SO₂ and CO₂ absorber tower.

The integrated tower concept is based on the proven dual loop tower that successfully has been used in many FGD applications. It holds a bottom section with a spray system separated from the top section. In the past the dual loop absorber has been used with packing so they can be installed to enhance CO₂ absorption in the top section.

From an economic point of view, the preferred use of CO₂ is for EOR in declining oil fields. Based on published data it is found that in the US several fields are active and that the average CO₂ injection is around 450,000 tpy. In this reference smelter the total amount of CO₂ captured is 576,000 tpy, which is in good agreement with an EOR scheme. To locate a new smelter near oil & gas fields has never been a major driver but perhaps the potential to store CO₂ in such fields may change the way future evaluations are conducted.

One concern that is associated with CCS is the consumption of electric power. This is considerable even by standards of aluminium smelters. There are two options, one that requires the purchase of 10-15% more power from the grid, and a second that means the power competes with the cells producing metal. In that case overall metal production is reduced by the equivalent amount of power used in the CCS unit.

Conclusions

Presented is the technical concept of application of carbon capture and sequestration (CCS) in conjunction with a primary aluminium smelter. While most people will agree that application of CCS in smelters presents challenges, near term international agreements may alter the course of thinking.

CCS is a collection of technologies that individually have been proven at commercial levels. As a combination they are not proven. If CCS is a candidate for application in primary aluminium smelters then this has to be tested in a scaled down version. Many technologies are tested or soon to be tested and time will tell which technologies will be the winners.

Schemes like EOR provide for positive returns on CCS. The combination of Dakota Gasification Company and EnCana Weyburn is an example that this concept can work on large scale.

Aluminium smelters provide for enough CO₂ to be able to effectively serve a regional EOR field. Even if this is over 300 km away, as is evidenced by the distance between Beulah and Weyburn.

CCS demand large amounts of energy. For application in aluminium smelters it will be critical to minimize the use of energy. Steam is generally not available and must be generated on-site. New technology is needed to extract the heat elsewhere where it needs to be dumped from the process.

In general, there are no major technical issues that withhold the application of CCS in primary aluminium smelters. That said, challenges exist and trials will reveal some of the details in the application. Albeit in different industries, CCS technology development takes place that directly is beneficial to this application as well. Good links with the power and oil & gas industries will be crucial to make CCS a success in smelters.

Finally, present work is a general view on the technical application of CCS. It is a topic that thus far is not reviewed in detail by the industry with the exception of a few peers. This paper is meant to contribute towards future discussions for the time that this concept is to be considered for the industry to make its contribution to a global effort. One such discussion should be on the detailed cost of applying CCS in primary aluminium smelters.

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