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MAGNESIUM: CURRENT AND ALTERNATIVE PRODUCTION ROUTES

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ABSTRACT

Magnesium is a light metal that used in structural applications, and also used as additive in chemical and metallurgical industries. The current dominant route for producing magnesium is via the Pidgeon process, a batch pyrometallurgical route that is energy-intensive and has high greenhouse gas emissions compared to other metal production routes. China currently dominates the world production because of the relatively low operating costs in that country. Australia does not currently produce magnesium, but the abundance of raw materials such as magnesite and dolomite, combined with the expected growth in magnesium demand associated with automobile lightweighting, have fuelled research and development in this area. This paper analyses the progress of magnesium production in the world, identifies problems and technical challenges associated with current and alternative technologies, and examines the prospects of establishing a magnesium industry in Australia.

Keywords: magnesium, primary production

INTRODUCTION

Magnesium, light metal has a density of 1783 kg/m^3 , which is two thirds of aluminium and one sixth of steel. The strength-to-weight ratio of magnesium is 158 kNm/kg , which is higher than 130 kNm/kg for pure aluminium. This high strength-to-weight ratio is the principal driver for the increasing demand of this metal. The purity required for commercial magnesium according to ASTM B92 (2007) for 9980A grade is a minimum of 99.80 wt% Mg, with impurities such as Ca, Al, Si, and Fe below 0.05 wt% each.

Magnesium has a number of applications, such as a light alloy in in the automotive industry (Mordike and Ebert, 2001), an alloying element in aluminium alloys (41% of US consumption in 2009), die casting (32%), steel desulphurisation (13%), and other applications as an industrial chemical (14%) (USGS, 2009). Aluminium industry utilizes magnesium as alloying ingredients to increase the strength, ductility and corrosion resistance of aluminium alloys (Slade, 2010). Magnesium's use in both aluminium and steel production strongly links its demand to these two other metal commodities.

The usage of magnesium has historically been limited by relatively high cost of production and associated energy costs. There have also been scientific issues around alloy development, in particular, increasing creep resistance for drive train applications and improving corrosion resistance. The primary production of magnesium has greatly increased in the past decade, as shown schematically in Figure 1. World production decreased in 2008 due to the global financial crisis, but the outlook for magnesium in the short to medium term is more optimistic. US and Canada dominated magnesium production during the 1990s, however, since the late 90s, the industrial revolution in China has seen that country become the main producer (Zang and Ding, 2001). This unprecedented growth in Chinese magnesium production was started by small operations growing simultaneously across the country which

take advantage of low operational cost, such as energy and labour costs. In general, electrolytic producers in the west have been replaced by Chinese pyrometallurgical production via the the Pidgeon process. The Pidgeon process is a relatively easy operation (i.e. does not require a highly trained work force or sophisticated engineering), versatile (i.e. easy to adjust production to meet demand) and only requires a small amount of capital cost compared to electrolytic processes. For example, the capital cost for Australia Magnesium (AM) Process using an electrolytic route was estimated to be \$10,000/tonne Mg, while the capital cost for the Pidgeon process was estimated to be \$3,000/tonne Mg in 2008 (Das, 2008).

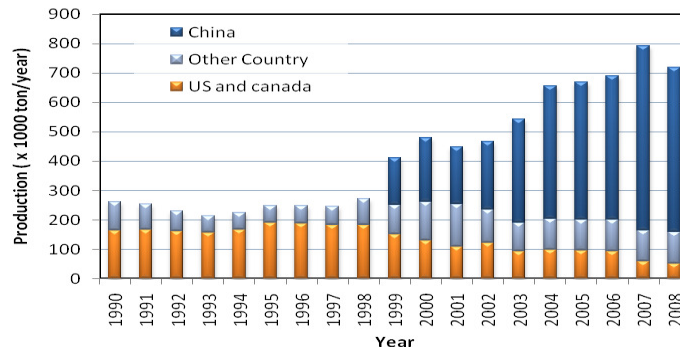


Figure 1. Primary Magnesium Production from International Magnesium Association (2010). The “Other Country” data before 1999 refers to any country besides US and Canada.

The Pidgeon process suffers from a low productivity, high labour requirement and high energy consumption, compared to other pyrometallurgical processes. The ferrosilicon reactant used in the process is also produced by an energy-intensive system. Thus, the Global Warming Potential (GWP) associated with the Pidgeon process is high; approximately 50% higher than aluminium and an order of magnitude higher than steel (Norgate and Rankin, 2007).

The desire to develop a more sustainable route for magnesium production has seen significant work around the world on developing new processes. In the 1990s, Australian developers looked to develop a new solvent extraction/electrolytic route in Queensland (Koenig *et al.*, 2002). Unfortunately, despite significant investment, the AM process was never commercialised. Since that time, Australian researchers have concentrated on developing a high temperature route to challenge the Pidgeon process, with Commonwealth Scientific and Industrial Research Organisation (CSIRO) concentrating on a carbothermic route (Brooks *et al.*, 2006, Prentice and Nagle, 2009). South African researchers at Mintek have also been developing a new pyrometallurgical route (Schoukens *et al.*, 2006).

PROCESSING ROUTES

In general there are two principal routes for producing magnesium. First, an electrolytic route in which liquid magnesium is won from magnesium chloride. The source of magnesium can be from sea water, brine, dolomite, magnesite, and carnallite. The other process involves reduction of magnesium ores by a reactant. A list of selected magnesium production routes are given in Table 1.

Electrolytic Processes

Electrolytic processes have dominated production magnesium from the 1970s to 1990s. In general, these processes include feed preparation, dehydration of magnesium chloride and electrolysis (Habashi, 1997). Figure 2 shows various process steps in electrolytic route which

are described in detail by Kipourous and Sadoway (1987). The feed preparation stage depends on the raw material. Magnesium oxide can be extracted from seawater by adding lime to form magnesium hydroxide. This is roasted to form caustic magnesium oxide. In the hydro magnesium route, magnesite is dissolved in hydrochloric acid forming a concentrated magnesium chloride solution.

Table 1. Selected Magnesium Production Processes

Process Route	Sources	Feed Preparation	Reaction	Temperature /Pressure
Electrolytic				
Dow process ¹	Brine/ seawater	Neutralization, purification, dehydration	Electrolytic $MgCl_{2(l)} \rightarrow Mg_{(l)} + Cl_{2(g)}$ Cathode: $2Cl^- \rightarrow Cl_2 + 2e$ Anode: $Mg^{2+} + 2e \rightarrow Mg$	T = 700 – 800 °C P = 1 atm
AM process ²	magnesite	Mining, leaching with HCl, dehydration		
IG Farben process ¹	Sea water/ brine	Neutralisation, prilling, dehydration chlorination		
Thermal Reduction Process				
Silicothermic ³	Dolomite FeSi	Calcination; FeSi making; pelleting	$MgO + CaO + FeSi = Mg_{(g)} + Ca_2SiO_{4(s)} + Fe_{(s)}$	T = 1160 °C, P = 13 – 67 Pa (1.2×10^{-4} atm)
Carbothermic ⁴	Magnesite, carbon	Calcination; pelleting	$MgO + C = Mg_{(g)} + CO_{(g)}$	T = 1700 °C P = 1 atm
Magnetherm ⁵	Dolomite, bauxite, FeSi	Calcination; FeSi making;	$2 CaO \cdot MgO + (x Fe) Si + n Al_2O_3 = 2CaO \cdot SiO_2 \cdot nAl_2O_3 + 2 Mg + x Fe$	T = 1550 °C P = 0.05 atm
Alumino-thermic ⁶	Dolomite Al scrap	Calcination	$4MgO_{(s)} + 2Al_{(s)} = 3Mg_{(g)} + MgAl_2O_{4(s)}$	T = 1700 °C P = 0.85 - 1 atm
Mintek ⁷	Dolomite, bauxite, FeSi, Al Scrap	Calcination	$2 CaO \cdot MgO + (x Fe) Si + n Al_2O_3 = 2CaO \cdot SiO_2 \cdot nAl_2O_3 + 2 Mg + x Fe$ $4MgO_{(s)} + 2Al_{(s)} = 3Mg_{(g)} + MgAl_2O_{4(s)}$	T = 1700 °C P = 0.85 atm

Reference:

¹(Habashi, 1997), ²(Jenkins *et al.*, 2009), ³(Mayer, 1944), ⁴(Brooks *et al.*, 2006), ⁵(Faure and Marchal, 1964), ⁶(Wadsley, 2000), and ⁷(Schoukens *et al.*, 2006)

There are two main Anhydrous routes for producing dehydrated magnesium chloride cell feed. These involve the Chlorination of magnesia (MgO) and the Dehydration of aqueous magnesium chloride. The IG Farben process includes mixing of magnesium oxide and charcoal prior to pelleting. The chlorination is carried out inside a brick-lined cylindrical shaft furnace where chlorine gas is passed from the bottom. Molten magnesium chloride is collected in the bottom of the furnace and transported to the electrolytic cells. In the dehydration of aqueous magnesium chloride route, seven process steps are required to achieve the anhydrous form.

Dehydration of magnesium chloride is energy intensive, but allows a more efficient electrolytic process and produces marketable chlorine. In contrast, partial dehydration (up to $MgCl_2 \cdot 2H_2O$), as in Dow process, requires less energy and less sophisticated preparation, but will result in consumption of the carbon anode and the chlorine produced is not marketable.

The AM process (Koenig *et al.*, 2002) was based around a solvent extraction route using ethylene glycol, as a means to produce anhydrous magnesium chloride feedstock from magnesite to a standard Alcan electrolytic cell. The complexity of the solvent extraction route partly explains the high capital cost associated with the process (Das, 2008).

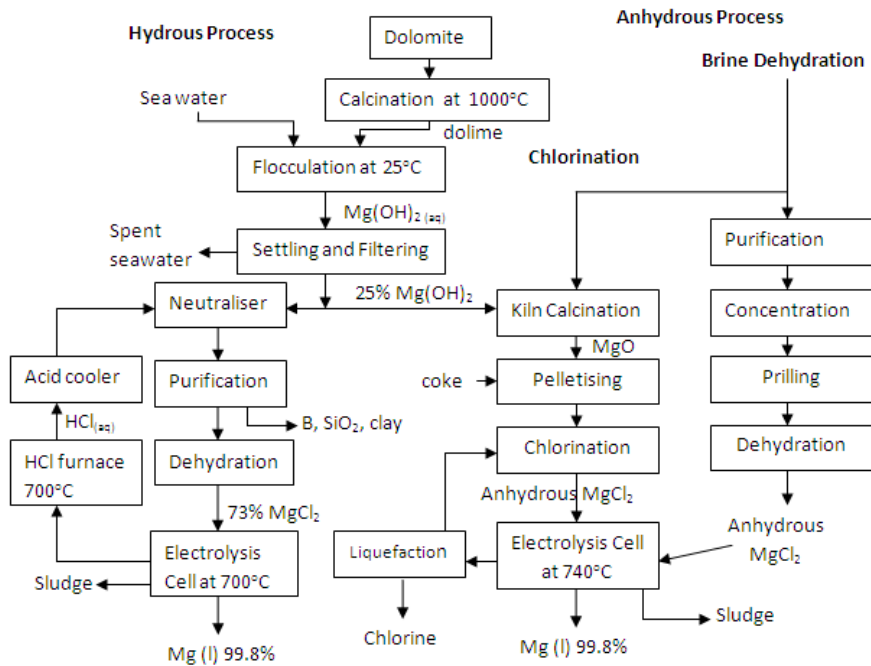
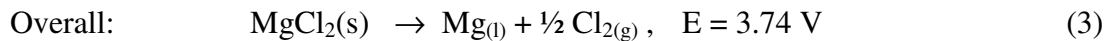
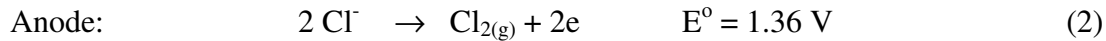


Figure 2. Flowsheet of Electrolytic Routes: Hydrus (Dow Chemical Process) and Anhydrous (IG Farben, Norsk Hydro, VAMI). (Kipouros and Sadoway, 1987)

Electrolysis of magnesium chloride dissolved in molten chloride salts occurs at 700 to 800 °C inside brick-lined vessels. For the anhydrous process, the electrolysis of magnesium chloride can be written as follows:



The cell designs, as detailed in Habashi (1997), varied between companies. While the theoretical energy usage in electrolysis of magnesium chloride is 7 kWh/kg Mg, the typical energy consumption of electrolytic cell varies from 10.5 to 13.2 kWh/kg Mg (Evans, 2007). The main cause of the loss energy is electrical resistance of the cell, which is the sum by resistances of cathode, anode, and electrolyte. The resistance of electrolyte is influenced by the properties of electrolyte and the distance between anode and cathode. Impurities in the feed also lower the cell performance by passivating the cathode surface which lowers the current efficiency. The impurities in the feed for IG Farben cell have a maximum contaminant level of 0.11% (0.1% MgO and 0.01% Fe).

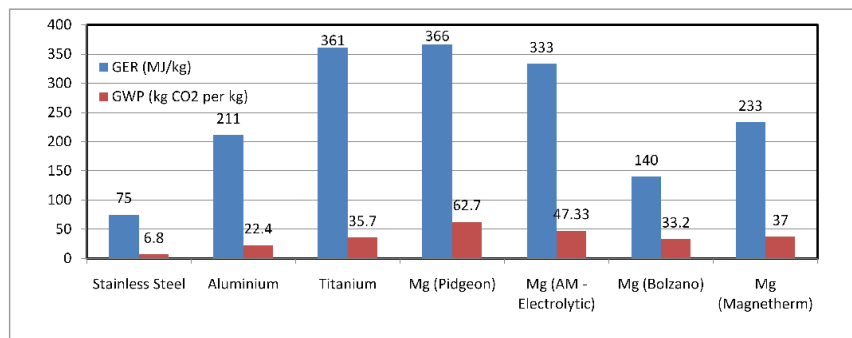


Figure 3. Comparison between Energy Requirement and Global Warming Potential of Metal Processes, especially Magnesium ((Cherubini *et al.*, 2008) and (Norgate and Rankin, 2007)).

The overall energy requirement estimated for the AM electrolytic process was 92.5 kWh/Mg, or 333 MJ/kg Mg ingot, as shown in Figure 3. The leaching and dehydration required 102.4 MJ/kg Mg or 30% from overall energy. While the energy usage for the electrolytic step was estimated to be 13.2 kWh/kg. The overall efficiency of electrolytic cell can be estimated to be 20.8% considering the electricity for electrolytic stage supplied by black coal (Ramakhrisnan and Koltun, 2004).

Pidgeon Process

The Pidgeon process is based on silicothermic reduction of magnesium oxide (Pidgeon, 1944), see Figure 4 for a flow sheet of the process. The calcination of dolomite takes place in rotary-kiln that operates at temperature ranges of 1000 to 1300 °C. Ferrosilicon is produced by the carbothermic reaction of quartzite in submerged electric arc furnace at 1600 °C. Calcined dolomite and ferrosilicon are mixed and briquetted prior to be placed in horizontal Ni-Cr stainless steel retort. At temperatures around 1160 °C and operating between 13 to 67 Pa, the reduction of calcined dolomite by ferrosilicon produces magnesium vapour. This reaction can be represented by

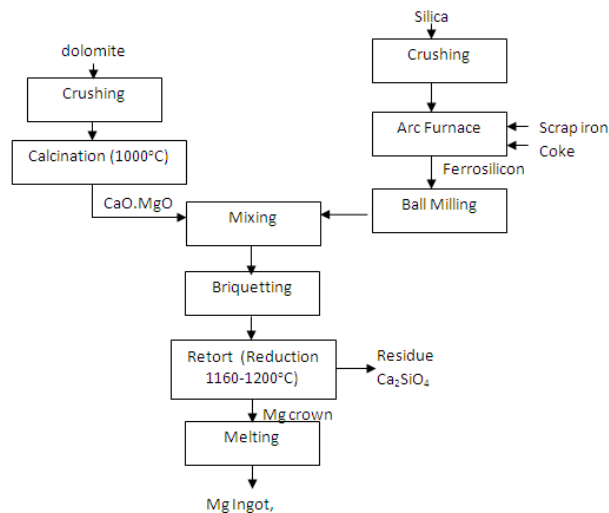
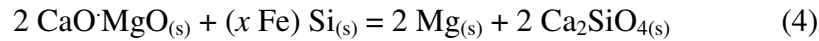


Figure 4. Schematic Flowsheet of the Pidgeon (Mayer, 1944)

The magnesium vapour condenses in a water cooled condenser unit outside the furnace. High purity magnesium can be obtained since the vapour pressure of potential impurities (Ca, Fe and Si) is low at these conditions. The low temperature of the process results in slow kinetics and poor heat transfer. The heat transfer problems limit the size of the reactor, and result in relatively low production rates. The typical operation per retort (reactor) is 20 kg magnesium per 8 hours of operation from a 128 kg charge (Ramakhrisnan and Koltun, 2004).

The Pidgeon process has a high energy demand, 366 MJ/kg Mg, compared to other common metals, as shown in Figure 3. The process suffers from excessive heat loss associated with the reduction process and ferrosilicon making (Cherubini *et al.*, 2008). Life Cycle Analysis calculations made by Ramakhrisnan and Koltun (2004) show that ferrosilicon production and reduction processes contribute to 31% and 50% of the energy consumption in magnesium production respectively. Ramakhrisnan and Koltun (2004) estimates the typical reduction process using a coal-fired furnace has an energy efficiency of only 12% whilst the estimated energy efficiency of ferrosilicon production in arc furnace is 51.5%.

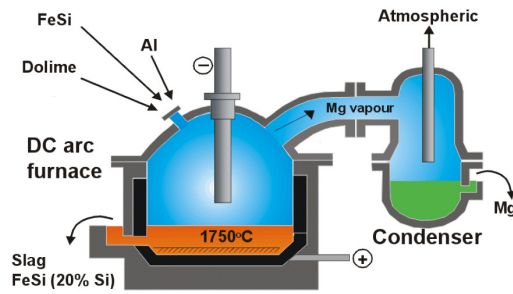


Figure 5. The Schematic of Mintek Process (Abdel-Latif and Freeman, 2008)

Mintek Process

The Mintek Process is a large scale batch silicothermic process operating at atmospheric pressure. It attempts to overcome the productivity and operational difficulties associated with an earlier large batch process called the Magnetherm process. Pechiney developed the Magnetherm process in the 1950s and operated at 1550 °C under vacuum. The vacuum conditions in the main reactor resulted in a number of practical problems. The process is currently not operational. Cameron *et al.* (1996) identified operating conditions that could avoid the difficulties associated with Magnetherm and productivity limitations of the Pidgeon process. A schematic of the Mintek process furnace is shown in Figure 5. The key to this process relies on the utilization of slag and aluminium, in addition to ferrosilicon, for the reduction of magnesium oxide at higher temperatures, 1700 to 1750°C. Thus, the vapour pressure of magnesium is much higher, at about 0.85 atm.

Table 2: Silicothermic process operating condition and impurities

Silicothermic Process	Pidgeon ¹	Magnetherm ²	Mintek ³
Operating Conditions			
Pressure (atm)	1.2×10^{-4}	0.05	0.85
Temperature (°C)	1100 – 1200	1550 – 1600	1700 – 1750
Productivity per furnace per day	50 kg	20 tonnes	100 tonnes
Indicated impurities in crude magnesium, (in wt%)			
Al	0.004	0.01	0.066
Si	0.010	0.05	0.281
Ca	0.005	0.005	0.385
Fe	0.007	0.005	0.250

Reference: ¹ (Mayer, 1944), ² (Faure and Marchal, 1964), ³ (Schoukens *et al.*, 2006)

This process has higher productivity than the Pidgeon process (see Table 2). However, the higher operating temperatures result in higher impurity levels due magnesium. Thermodynamic calculations predict a magnesium purity of 97.86 wt% for the Mintek process compared to 99.68% for the Pidgeon process (Wulandari *et al.*, 2009). This increased impurity level necessitates a subsequent refining stage, that adds to the process operating and capital costs. The problems associated with high temperature silicothermic processing are currently being studied by the authors.

Alternative Routes

The Solid Oxide Membrane (SOM) process (Pal *et al.*, 2001) can be alternative to electrolytic process. In this process, reduction of magnesium oxide dissolved in fluoride-based electrolytes (MgF₂-CaF₂-MgO) is carried out by passing electric current at 1150 to 1300 °C.

When electrical current is applied, magnesium oxide dissociates into magnesium and oxygen. Oxygen ions are pumped out through Yttrium Stabilised Zirconia (YSZ) membrane to the anode, as illustrated in Figure 6. The magnesium vapour evolves at the cathode and condenses in a separate chamber. The reported energy demand is 10 kWh/kg Mg.

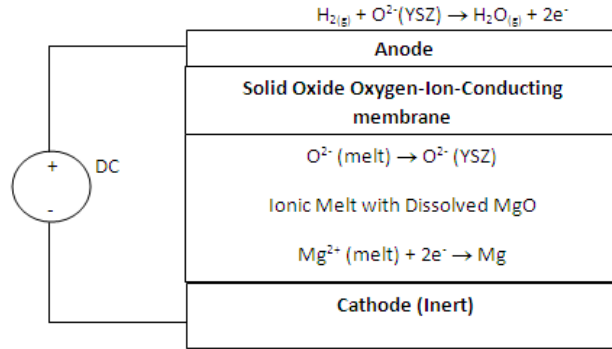
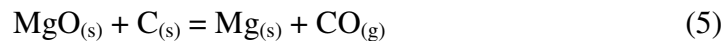


Figure 6. The SOM Process for Magnesium Production (Pal *et al.*, 2001)

There are several scientific challenges in the SOM process. The selection of fluxes is crucial since the YSZ membrane should not dissolve into the MgF_2 - CaF_2 - MgO flux. The leakage of current in the cell because of oxygen impurity in the cell is also an issue. The engineering challenges regarding thermal shock and mechanical failure is a problem since the membrane used is the most expensive parts in SOM cell (Krishnan *et al.*, 2005).

Carbothermic routes are an alternative to both silicothermic and electrolytic process. Carbothermic magnesium routes are currently being developed by CSIRO in Australia and by researchers in the USA. The process involves carbon to reduce magnesium oxide which produces magnesium vapour and carbon monoxide gas above 1500 °C. The overall reaction can be written as:



The advantages of the process are cheaply reducing agent and the potential for continuous operation. The carbon source is readily available, such as from petroleum coke, charcoal, and char. However, the shortcomings come from the reverse reaction of magnesium vapour and carbon monoxide gas as the mixture is cooled down. This result in fine magnesium powder contaminated with magnesium oxide.

The developments of carbothermic have concentrated to overcome the reversion issue on two process routes: The “quench” route and the “solvent route”. In the “quench” route, magnesium and CO are generated are rapidly quenched and recovered. The quenching engineering system operates at 1000000°C/s by using a Laval nozzle. The fundamentals of this process have been established (Brooks *et al.*, 2006, Prentice and Nagle, 2009). Figure 7 illustrates the schematic of carbothermic route. The system is currently being scaled-up to pilot plant operation (CSIRO, 2010). Like the high temperature silicothermic processes, the greater productivity of the carbothermic process will also be associated with increased impurities in the primary stage that require further refining (Brooks *et al.*, 2006).

The ‘solvent’ route is another carbothermic process (Brooks *et al.*, 2006). In this route, agglomerated feed is fed into the reduction phase containing a molten metal solvent in which magnesium formed from reduction dissolved in the molten metal while the CO gas librates. The great advantage of this route is the separation of magnesium from CO gas, but limited

choice of metal is one of the scientific issues facing this route. The process is currently very theoretical and requires significant work before it can be developed.

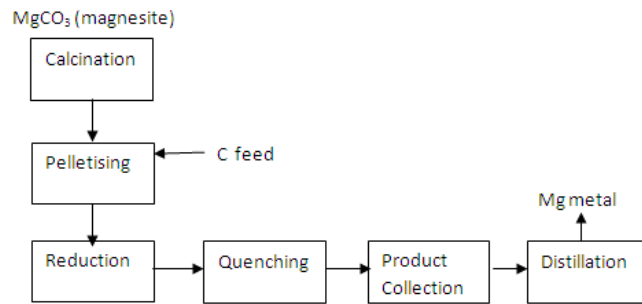


Figure 7. A Schematic Flow Sheet of Quenching Route of Carbothermic (Brooks *et al.*, 2006)

Aluminothermic routes, such as Heggie-Iolaire Process (Wadsley, 2000), offer attractive choice over other production routes. Aluminium is a stronger reducing-agent compared to ferrosilicon and carbon. The aluminothermic reaction can be written by:



This process would only be possible if only there is a cheap source of aluminium resources available. Through a Life Cycle approach, the process is also unattractive because of the energy required to provide aluminium is also high, which is 211 MJ/kg Al (Figure 3).

MAGNESIUM IN AUSTRALIA

Australia has abundant magnesium mineral resources such as dolomite and magnesite, spread around the continent. The largest identified source is an ore body northwest of Leigh Creek (South Australia) with 235 Mt of magnesite. The second largest inventory is the Kunwarara deposit, Queensland, in the form of low iron nodules of cryptocrystalline magnesite with the identified resources at 87 Mt of “run of mine” ore. It is currently mined for refractory purposes with a production of 540,000 tonnes/year (Geoscience Australia (2002).

Australia currently does not produce magnesium, although there have been several attempts to establish magnesium plants in the late 90s. Australian Magnesium Corporation had planned to build the biggest up to magnesium smelter using electrolytic route (AM process) in Stanwell, Queensland, but closed before operation started (Chessel, 2003). The South Australia Magnesium (SAMAG) planned to build magnesium plant in South Australia utilising the magnesite deposit at Leigh Creek sediment to provide raw material in a Dow process. LaTrobe Magnesium also plan to build 100,000 tonnes per annum smelter using the Pidgeon process technology from brown coal fly-ash produced from existing power generation in Latrobe Valley, Gippsland, and currently in feasibility study phase (Anonymous, 2008). Typical lignite coal fly-ash has MgO content of 3 to 10% and CaO content of 15 to 40%.

The impact of Chinese magnesium output growth is the main reason why there is no magnesium project completed. The high energy requirements are also a serious challenge in establishing a magnesium production facility in Australia.

DISCUSSION

Magnesium market will steadily increased because of its attractiveness as weight reduction material in automotive and aerospace industry. Weight reduction through alloy substitution by magnesium can reduce fuel consumption and lower Green House Gas emissions

(Tharumarajah and Koltun, 2007). However, the high energy usage in magnesium primary production may offset any environmental advantage.

As shown in Figure 3, the energy consumption of magnesium production is high compared to other metal production routes. It is 50% higher than aluminium, and six times higher than stainless steel. The energy consumption for producing metal from ore is strongly linked to the chemical stability of mineral being processed, availability and the richness of ore, and process route (Brooks and Subagyo, 2002). Beside the abundance of the ore, magnesium concentration in the ore is relatively high. Magnesium concentration in seawater is 0.13%, while the concentration in dolomite and magnesite are 13% and 28.8% accordingly.

Table 3. Chemical Stability of Selected Oxide

Reaction	Gibbs Free Energy (ΔG) at 25 °C (MJ/kg metal)	Enthalpy Formation (ΔH) at 25 °C (MJ/kg metal)
$2 \text{ Mg} + \text{O}_2 \rightarrow \text{MgO}$	-23.42	24.76
$2 \text{ Al} + 3/2 \text{ O}_2 \rightarrow \text{Al}_2\text{O}_3$	-29.3235	31.2
$2 \text{ Fe} + 3/2 \text{ O}_2 \rightarrow \text{Fe}_2\text{O}_3$	-6.66	7.30

The chemical stability of an oxide compound can be reflected by the Gibbs free energy while the enthalpy formation determines the minimum energy requirement of a process. As seen in Table 3, the Gibbs energy of magnesium oxide is lower than iron oxide, which indicates that magnesium oxide is more stable and requires more energy to extract the metal. The difference between the theoretical energy requirements and the actual energy usage reflects the different aspects of the process route. The energy efficiency of ferrosilicon electric arc furnace is only 51.5% while the efficiency of coal fired furnace is 12%. The Pidgeon process urgently needs improvement in order to reduce energy consumption and greenhouse gas emissions. Improvement that has been proposed includes utilisation of cleaner energy source (e.g. natural gas or producer gas) and integrating small smelters to improve raw material and energy efficiency (Aghion and Bartos, 2008).

The electrolytic process has a lower Global Warming Potential compared to the Pidgeon process (47.3 kg CO₂/kg Mg to 62.7 kg CO₂/kg Mg), but suffers from low productivity and high capital costs. The development of new high productivity pyrometallurgical processes by CSIRO and Mintek may provide the route by which Western countries, such as Australia, could develop an environmentally and economically acceptable route to Magnesium production.

CONCLUSIONS

The light weighting of transportation and alloy development will continue be the driving force of the increasing demand of magnesium metal. Magnesium production consumes greater energy than other metal production routes due to its oxidechemical stability. In particular, the Pidgeon process as the dominant production route in the world has high energy consumption which leads to high a Global Warming Potential. Thus, technology improvement is essential to achieve lower energy usage and a sustainable future. New processes such as Carbothermic and the Mintek process are high productivity alternatives to the existing technologies that still require further development.

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