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MAGSONIC™ CARBOTHERMAL TECHNOLOGY COMPARED WITH THE ELECTROLYTIC AND PIDGEON PROCESSES

Leon H. Prentice[1]; Nawshad Haque[1]
[1]CSIRO Process Science and Engineering; Box 312; Clayton South, VIC, 3169, Australia

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Abstract

A broad technology comparison of carbothermal magnesium production with present technologies has not been previously presented. In this paper a comparative analysis of CSIRO's MagSonic™ process is made with the electrolytic and Pidgeon processes. The comparison covers energy intensity (GJ/tonne Mg), labor intensity (person-hours/tonne Mg), capital intensity (USD/tonne annual Mg installed capacity), and Global Warming Potential (GWP, tonnes CO_2-equivalent/tonne Mg). Carbothermal technology is advantageous on all measures except capital intensity (where it is roughly twice the capital cost of a similarly-sized Pidgeon plant). Carbothermal and electrolytic production can have comparatively low environmental impacts, with typical emissions one-sixth those of the Pidgeon process. Despite recent progress, the Pidgeon process depends upon abundant energy and labor combined with few environmental constraints. Pressure is expected to increase on environmental constraints and labor and energy costs over the coming decade. Carbothermal reduction technology appears to be competitive for future production.

Introduction

Magnesium is the lightest of the structural metals and has significant potential as an important material for future manufactured products, most particularly in the automotive industry for the production of light-weight and more fuel-efficient vehicles. However, difficulties in extracting magnesium from its sources – whether brines, dolomite, magnesite, fly ash, etcetera – have so far limited its production and made it more expensive than other metals, most notably aluminum. Furthermore, present methods of production are generally perceived as excessively capital-intensive or environmentally damaging [1, 2].

A new method of production, based on carbothermal reduction technology with supersonic quenching, has been developed by Australia's Commonwealth Scientific and Industrial Research Organisation (CSIRO). This technology is approaching the point of significant scale-up and commercialization, and promises advantages over existing technologies. Process development over the last 2-3 years has produced the information required for a meaningful comparison with existing technologies, although further proof and refinement of the data will come with increased development.

In this study the MagSonic™ process is compared with the electrolytic and Pidgeon (Silicothermic) processes. Comparisons may be made on the projected energy intensity, labor intensity, capital intensity, and Global Warming Potential. Other technologies, either historical or developmental, are briefly discussed, but no formal comparisons are made. While these are not the only useful indicators of comparison, they can be assessors of the potential new competitor in global production.

Magnesium Production Technologies

Electrolytic Production

Historically, the electrolytic process was dominant from the start of commercial production up until the 1990s. Significant production facilities were constructed around the world, the largest of which was Dow's plant at Freeport, TX. Presently, magnesium is produced using the electrolytic process in the United States, Russia, Ukraine, and Israel, with potential start-up in China.

While there are a number of variations of the electrolytic process, all now electrolyze anhydrous magnesium chloride or carnallite. The overall reaction is:

$$MgCl_{2(l)} \rightarrow Mg_{(l)} + Cl_{2(g)} \qquad (1)$$

In the carnallite process, the potassium chloride (KCl) is chemically a spectator, and is recycled. The Dow process electrolyzed the dihydrate $MgCl_2.2H_2O$; the waters of hydration were 'fizzed off' in the electrolysis cell [3].

High purity molten Mg is tapped from the cells and cast into ingots. The details of the electrolytic process have been described in detail elsewhere [3-5].

Pidgeon (silicothermic) Production

The Pidgeon Process is named after Lloyd Montgomery Pidgeon, who developed the process in the 1940s [6-8]. The process reduces magnesia (in the form of calcined dolomite or 'dolime', $CaO.MgO$) with silicon (in the form of ferrosilicon, $FeSi$). Various species are essentially spectators, but the overall reaction can be described by:

$$2(CaO.MgO)_{(s)} + FeSi_{(s)} \rightarrow 2Mg_{(g)} + 2CaO.SiO_{2(s)} + Fe_{(s)} \qquad (2)$$

The production of magnesium is by batch reaction of briquettes (often with a flux such as CaF_2 added), with approximately 40 to 70 kg of magnesium produced from each retort over its 5-10 h cycle. Various fuels may be used to provide heat – the most common fuels are brown coal, natural or coal gas, and electricity from coal or hydroelectric power. High-purity 'crowns' are produced, which are then re-melted to produce Mg ingots.

Carbothermal (MagSonic™) Production

In carbothermal production, magnesia is reduced directly with carbon in the high-temperature gas-solid reaction:

$$MgO_{(s)} + C_{(s)} \leftrightarrow Mg_{(v)} + CO_{(g)} \qquad (3)$$

The process has been described comprehensively by Brooks *et al* [9, 10]. The reverse arrow in the reaction above indicates that the back-reaction is rapid and spontaneous upon cooling; such reversion may be minimized by very rapid cooling. CSIRO's MagSonic™ process, under development in Australia since 2003, achieves minimal reversion by cooling the reaction products (gases) at up to $1x10^6$ °C.s^{-1} by supersonic acceleration through a Laval nozzle. The magnesium powder is purified and cast into ingots, while the carbon monoxide may be burnt to recover additional energy. The simplicity of direct reaction with carbon is apparent – carbon is plentiful, cheap, and potentially renewable (e.g. biochar) – and the only significant process waste is stoichiometric CO_2.

Other Production Methods

Bolzano. The Bolzano process has been modified and expanded and is now operated by Rima in Brazil, producing around 22,000 tonnes Mg per year [11]. It uses Pidgeon chemistry, but in extremely large vertical retorts, producing up to 2,000 kg per ten-hour cycle [11]. While a formal comparison with the Bolzano process is not part of the present work, Rima's claim to be a 'green producer' is supported by Cherubini *et al*'s analysis [12] of an energy intensity (Gross Energy Requirement) around 140 GJ/tonne Mg – half the electrolytic figure and around 60% lower than the Pidgeon process. A low environmental impact is maintained by the use of charcoal and hydroelectric power [11].

Magnétherm. The Magnétherm process – a continuous variation of the Pidgeon chemistry – is no longer in use. A similar process developed by Mintek [13, 14], operating at higher pressure, appears to have not been commercialized. However, Cherubini *et al*'s [12] analysis indicated that the energy intensity of the Magnétherm process was between the Bolzano and Electrolytic processes, at around 233 GJ/tonne Mg.

Solid Oxide Membrane (SOM). The SOM process has been under development by Boston University for some time, and has reported some experimental and scale-up success [15-17]. It is based on electrolysis of MgO in a fluoride-based ionic flux. Das [18] reported in 2008 that the SOM team had achieved up to 100 g.day^{-1} of Mg production. However, more recent work [17] discusses issues in membrane stability, and the future development of the process is uncertain. While Das [18] reported a very attractive commercial parameters – cash operating cost around USD 0.53 per pound, capital intensity around USD 1,345 per tonne installed capacity, and labor intensity only 1.35 person-hours per tonne – the small scale and development uncertainty in the SOM process has excluded it from the present comparison.

Technology Comparison

Energy Intensity

The most recent data presented on Chinese magnesium production are from the 2011 IMA (International Magnesium Association) meeting, reported by Brown [11]. He reports Chinese Magnesium Association (CMA) data of average energy requirements 'around 5 tce/t' (tonnes of coal equivalents per tonne magnesium) and CO_2 emissions of '~12-17' tonnes/tonne Mg. (See below for discussion on emissions.) Coal's calorific value is around 23 MJ/kg [19], hence CMA's energy intensity estimate is around 115 GJ/tonne Mg. Also from 2011, Feng *et al* [2] report that 'the comprehensive energy consumption of 1 [tonne] of primary magnesium... decreases from 360 GJ/t Mg to 297 GJ/t Mg between the year 2005 and 2009.' These figures compare with Du *et al*'s 280 GJ/tonne Mg [19], Cherubini *et al*'s 366 [12], and Ramakrishnan and Koltun's 354.5 [20]. Although detail of CMA's study has not been investigated by the present authors, it appears that the CMA has focused exclusively on the Mg reduction step, and their figure is therefore optimistic. Du *et al* [19], for example, do not take into account *FeSi* production, and hence arrive at a low estimate. (Ramakrishnan and Koltun [20] estimate *FeSi* production as requiring 113.5 GJ/tonne Mg. They also estimate that the energy intensity of the reduction step alone is 181.4 GJ/tonne Mg, which makes CMA's figure reasonable only for this part of the process.) The true overall figure likely remains around two to three times CMA's figure: Feng *et al* [2] are likely accurate at around 300 GJ/tonne Mg. Given present uncertainty and variations in process details, this figure may be only accurate to within 20%.

The gross energy requirement of the electrolytic process has been described in detail previously [12, 20-22]. Present research into further developing the electrolytic process is limited; developments remain either proprietary or focus on existing process routes [23, 24]. Technology advancement is unlikely to further reduce the estimate of around 270 GJ per tonne Mg. The estimate may be ±10%.

Data presented by the present authors in 2010 [21] disclosed that MagSonic™ carbothermal reduction is expected to require around 56 GJ/tonne Mg. Around 60% of the energy required is for the reaction step, for which an electrically-powered reactor has been estimated at 50% efficiency; the remainder is mostly vacuum systems, calcination, and purification. The data for the reactor are supported by laboratory operation of a 50 kW induction furnace over more than 200 experiments. The most significant advantages of the carbothermal route are the very simple feed preparation (calcination and briquetting), direct reaction, minimal wastes, and continuous operation. Given the present scale of the process, energy intensity estimates may be ±50%.

The energy intensity data are summarized in Table I below.

Table I: Energy Intensity Comparison

Process	Energy Intensity (GJ per tonne Mg)
Pidgeon	300 (±60)
Electrolytic	270 (±27)
MagSonic™	56 (±28)

Global Warming Potential

Foundational work on the life cycle analysis of the electrolytic (and Pidgeon process) was undertaken by Ramakrishnan and Koltun [20, 22, 25]. Their work was updated and expanded by various authors more recently [12, 19, 26]. These data were combined with the present authors' analysis in a comparison with the carbothermal MagSonic™ process in 2010 [21].

Overall, the data showed that the Pidgeon process in China had an environmental impact, measured in terms of Global Warming Potential (GWP), of around 42 tonnes CO_2-equivalent per tonne of Magnesium produced [12, 20]. (The data are obviously variable, and significant differences exist for different producers depending on the exact technologies used.) Ramakrishnan and Koltun's [20] estimated range is 37-47. Du *et al* [19] cite a figure of 27 tonnes CO_2-eq per tonne Mg, but do not include the impact of coal combustion, which may add between 16 and 40 to this figure, depending on efficiency and type of coal. The CMA's estimate of '~12-17' tonnes CO_2 per tonne Mg appears to focus solely on CO_2 emissions from the Mg reduction step, for which Ramakrishnan and Koltun [20] estimated 15.9 tonnes CO_2 *equivalents* per tonne. Feng *et al*'s data [2] appear more comprehensive, detailing a GWP of around 28 tonnes CO_2-eq per tonne Mg for the Pidgeon process in China in 2009. They report that the reduction from 2005 to 2009 is due primarily to improved efficiency, particularly in the 'coal gasification process and exhaust heat recovery in [the] reduction process.'

The average of Feng *et al*'s 2011 [2] data and 'base case' processes from 2005 leads to the overall comparison shown in Table II below. Variation in the Pidgeon process estimate is largely due to process variation, whereas the GWP of the electrolytic and MagSonic™ processes depends primarily on the source of electricity. With hydroelectric power, the MagSonic™ process can have an environmental impact as low as 6 tonnes CO_2-eq per tonne Mg.

Table II: Global Warming Impact Comparison

Process	GWP (tonnes CO_2-eq per tonne Mg)
Pidgeon	34 (±8)
Electrolytic	17 (±8)
MagSonic™	14 (±8)

Capital and Operating Cost

Data for capital and operating costs are tentative, variable, and highly dependent on location. While this paper reviews data from a number of sources, the final analysis is provided in US dollars for western construction (e.g. in Australia).

Only two Greenfield electrolytic plants have been built – or partially built – to the authors' knowledge, in the last two decades. Capital cost data are not readily available for the Magnola project [27], but data exist for the Australian AMC project, although it collapsed prior to completion. The funding estimate for that project was AUD 1.5bn for a plant to produce 90,000 tonnes Mg per year. Adjusting for inflation and using the long-term exchange rate of 0.8 USD/AUD leads to an installed capital cost around USD (2010) 16,667 per annual tonne Mg. This is similar to figures cited by Das [18]. Given paucity of data and time since estimates were made, an error estimate of 15% may be appropriate.

While the Pidgeon process (as presently operated) would be unfeasible for Australia – see below – it may be worthwhile to estimate its capital cost should such a plant be constructed. The capital cost of the CVM Minerals plant in Malaysia is a useful benchmark: its capital intensity is around USD 2,650 to 3,400 per annual tonne Mg [28]; a value somewhere around USD (2010) 3,250 may be reasonable for Australia. Comparable data on the Pidgeon process in China are difficult to obtain, but are likely lower. At the high end Das [18] also suggests USD (2010) 3,250. (One extremely low estimate is China Magnesium Corporation's Prospectus [29], which cites CNY 3,355 (approximately USD 500) per tonne installed capacity. This figure has not been included in the present analysis, which focuses on western production. Again, paucity of data suggests errors may be around ±20%.

Work by the present authors on the MagSonic™ carbothermal process results in a capital cost of around AUD 210m for a plant producing 30,000 tonnes Mg per year [30]. This equates to USD (2010) 5,600 per annual tonne Mg. The overall breakdown in cost is similar to that of Odle and McClaine [31], where around 25% of the capital cost is reactors, followed by the vacuum system (approximately 17%) and calcining plant (approximately 16%). The initial estimate is only to ±50% confidence.

There have also been literature analyses of the carbothermal process, all of which arrive at lower capital costs. Donaldson and Cordes [32] estimated an installed capital cost of USD (2005) 3,225 per tonne capacity, or USD (2010) 3,900. Odle and McClaine's 2007 study [31] estimated USD 4,500 (based on 90,000 tonnes per year capacity), or USD (2010) 5,040. Das [18] cites Odle an McClaine's data, but does not include contingencies and other costs, arriving at a capital cost of USD 221m for 90,000 tonnes Mg per year, or around USD (2010) 3,000 per tonne installed capacity. These estimates are included as literature values in Table III below.

Table III: Capital Intensity Comparison (western location, 2010)

Process	Capital Intensity (USD per annual tonne Mg capacity)
Pidgeon	3,250 (±650)
Electrolytic	16,667 (±2,500)
MagSonic™	5,600 (±2,800)
Carbothermal (literature)	4,000 (±1,000)

Operating costs are even more location and process-specific, so no detailed comparison is presented here. However, indicative comments and results are discussed below.

Odle and McClaine's 2004 work [33] produced an economic model for the carbothermal process: "the model predicts operating costs between about $0.23 and $0.55 a pound". Adjusting for inflation leads to an estimate of USD (2010) 0.49 (±0.20) per pound. Their paper focused on the structure and rationale of the model, using limited experimental results. Their later study [31] stated a cash cost "as low as $0.30 per pound" (USD (2010) 0.34 per pound). These figures compare with Donaldson and Cordes' estimates [32] of cash cost of production around USD (2010) 0.51 per pound Mg. Excluding capital, Das [18] estimates a cash cost of production via carbothermal reduction around USD (2010) 0.70 per pound.

Developments in MagSonic™ technology also allow for an operating cost estimate, of USD (2010) 1.19 per pound Mg. This figure is obviously significantly higher than those suggested previously [18, 31]. Part of the variation is due to electricity, of which around 15.61 kWh per kg Mg is required [21]. Global electricity prices vary significantly, but assuming USD 0.10 per kWh, the cost of electricity alone is USD 0.71 per pound Mg. It is likely that the cost of energy contributes to the variation in estimates in the literature for the carbothermal operating cost.

The operating cost of the electrolytic process is likewise highly dependent on the cost of electricity, and no meaningful comparisons can be made. A nominal figure somewhere around USD (2010) 0.98 per pound [18] seems reasonable.

In contrast to the carbothermal and electrolytic processes, the Pidgeon process relies primarily on labor (and to a lesser extent ferrosilicon) for its operating cost. Das [18] estimates USD (2010) 0.98 per pound in China, compared with 1.09 in a western location.

Labor Intensity

The labor intensity of the Pidgeon process has been frequently acknowledged [18, 31, 34], but exact figures are rarely found in the literature. Estimates may be made from scale and staffing figures, where available. The 'best case' scenario may be the recent construction of the Perak Magnesium Smelter, operated by CVM Minerals in Malaysia. Their plant is reported to employ 398 people for 15,824 tonnes per year of production [28], which at 1600 person-hours per year per worker equates to approximately 41 person-hours per tonne of production. A 'worst case' scenario may be the 2010 prospectus from Australia's China Magnesium Corporation, which expects labor costs of approximately CNY 956 per tonne [29]. At USD 1.36 per hour [35], this equates to around 105 person-hours per tonne. This latter figure is likely to be more common for the established plants that contribute the bulk of production. An average figure of around 90 person-hours per tonne appears reasonable for the Pidgeon process in China, with an estimated 20% error.

By contrast, CSIRO's work on the MagSonic™ process estimates a labor requirement of 5.8 person-hours per tonne Mg. This is largely due to the simplicity of the process and its likely capability of continuous operation. Given the ongoing development of the process, error is estimated as ±50%. Labor requirements for the electrolytic process are difficult to obtain, but estimates based on the size of the process plant and employment figures for producers suggest around 9-15 person-hours per tonne Mg may be reasonable.

These data are summarized in Table IV below.

Table IV: Labor Intensity Comparison

Process	Labor Intensity (person-hours per tonne Mg)
Pidgeon	90 (±18)
Electrolytic	12 (±3)
MagSonic™	6 (±3)

It is evident from the above analysis that construction of a Pidgeon process plant with its present labor intensity requirements would be prohibitive in countries with medium to high labor costs. In Australia, for example, labor costs may be estimated at around USD (2011) 35 per person-hour [36], rendering the Pidgeon process uneconomic.

Conclusion

Continued work by CSIRO on the MagSonic™ carbothermal production route for magnesium, combined with data available for competing technologies, has allowed for a comparison to be made. The results highlight the energy and environmental advantages of the carbothermal production of magnesium and indicate that the process is likely to be competitive on a cash operating cost basis. While its capital cost is likely higher than the Pidgeon process, labor intensity is very much lower. The lack of electrolytic construction is explained by its high capital cost, whereas the MagSonic™ process is likely to be competitive for Australian production.

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