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Life cycle assessment of gas atomized sponge nickel for use in alkaline hydrogen fuel cell applications

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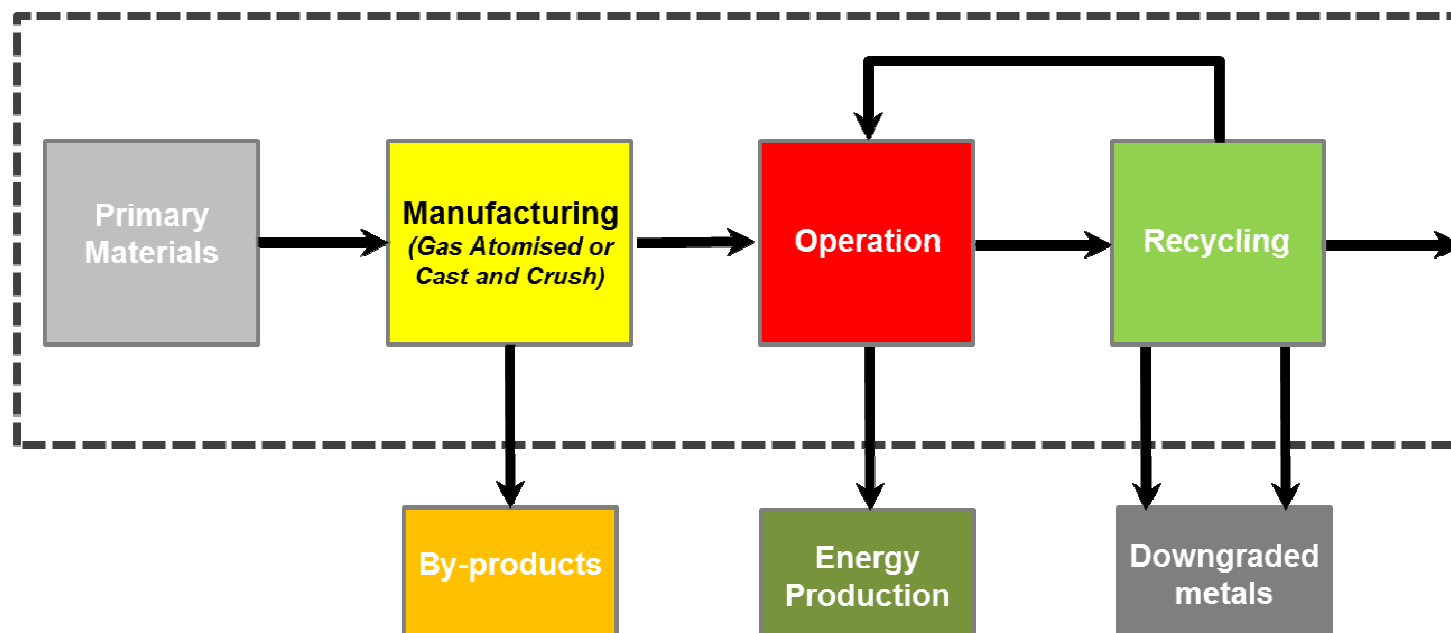
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Complete Life Cycle of Sponge Nickel

applications

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This paper presents a cradle-to-grave comparative Life Cycle Assessment (LCA) of new gas atomized (GA) sponge nickel catalysts and evaluates their performance against the both cast and crush (CC) sponge nickel and platinum standards currently used in commercial alkaline fuel cells (AFC). The LCA takes into account the energy used and emissions throughout the entire life cycle of sponge nickel catalysts - ranging from the upstream production of materials (mainly aluminium and nickel), to the manufacturing, to the operation and finally to the recycling and disposal. Through this assessment it was found that the energy and emissions during the operational phase associated with a given catalyst considerably outweigh the primary production, manufacturing and recycling. Primary production of the nickel (and to a lesser extent dopant materials) also has a significant environmental impact but this is offset by operational energy savings over the electrode's estimated lifetime and end of life recyclability. From the results it can be concluded that higher activity spongy nickel catalysts produced by gas atomization could have a significantly lower environmental impact than either CC nickel or platinum. Doped GA sponge nickel in particular showed comparable performance to that of the standard platinum electrode used in AFCs.

Keywords: Sponge (Raney) nickel • Gas Atomization (GA) • Platinum electrode • Alkaline Fuel Cell (AFC) • Cast and Crush (CC) • Life Cycle Assessment (LCA)

List of abbreviations

<i>AD</i>	<i>Abiotic Depletion</i>
<i>AFC</i>	<i>Alkaline Fuel Cell</i>
<i>AP</i>	<i>Acidification Potential (kg SO₂ eq.)</i>
<i>APU</i>	<i>Auxiliary Power Units</i>
<i>CC</i>	<i>Cast and Crush</i>
<i>CHP</i>	<i>Combined Heat and Power System</i>
<i>CML 2001</i>	<i>Institute of Environmental Sciences, Lieden</i>
<i>EDIP97</i>	<i>Environmental Design of Industrial Products</i>
<i>GA</i>	<i>Gas Atomisation</i>
<i>GABI</i>	<i>Software by PE international for lifecycle assessment</i>
<i>GER</i>	<i>Gross Energy Requirement (MJ)</i>
<i>GHG</i>	<i>Green House Gas</i>
<i>GWP</i>	<i>Global Warming Potential (kg CO₂ eq.)</i>
<i>LCA</i>	<i>Life Cycle Assessment</i>
<i>PEM</i>	<i>Polymer Electrolyte Membrane</i>
<i>PSD</i>	<i>Powder Size Distribution</i>
<i>SOFC</i>	<i>Solid Oxide Fuel Cell</i>

1. Introduction

Over the past twenty years fuel cell technologies have come to be regarded as one of the most promising alternative power sources due to their potential for high efficiency electricity generation and low environmental impact [1-4]. However, more recently with increasing legislation and enhanced environmental awareness efforts have been made to subject various fuel cell systems and applications to life cycle assessment (LCA). These assessments have embraced a wide range of aspects including the systematic development of the necessary tools for life cycle assessment of fuel cell powered vehicles as demonstrated by Contadini *et al.* [5] who combined the Fuel Upstream Energy and Emission Model (FUEEM) with academic/manufacturer literature and the work of Peht [6, 7], which addressed both methodological and environmental aspects with regard to Solid Oxide Fuel Cells (SOFC). In addition, such investigations have also concerned themselves with the fundamental question of fuel sources for the next generation of automotive propulsion [8, 9] including various biofuels [10] as part of this holistic approach towards the environmental impact of fuel cell technologies.

In contrast other researchers have focussed more on specific fuel cell types or applications including SOFC [11, 12], polymer electrolyte membrane, (PEM) [13] microbial [14] and fuel-cell based auxiliary power units (APUs) [15]. Alkaline Fuel Cell (AFC) type fuel cells have themselves been the subject of a comprehensive LCA by Staffell and Ingram [16] who assessed the impact of including an AFC fuel cell as part of a domestic combined heat and power (CHP) system. Their results showed that production of the AFC stack is relatively insignificant when compared to the other components of the CHP system, but that the biggest environmental impact was from the sulphur dioxide (and other generated respiratory

Overall, it was concluded that improvements to the nickel catalysts used - in terms of increased activity and longevity – and better design to ease of disassembly and recycling are key to improving environmental performance of AFC and fuel cells in general.

The use of Raney or Sponge nickel [17, 18] has been investigated in a number of fuel cell applications including as a basis for the electrodes in both AFC [19-21] and SOFC [22] and low temperature AFCs are attracting renewed interest [23] due to the potential of low material costs coupled with high system efficiencies. One of the driving factors is an economic benefit based on the potential to replace platinum and palladium by nickel-based electrodes. This has related environmental benefits, due to the scarcity of the noble metals in the Earth's crust the energy requirements and emissions associated with their mining and concentration are orders of magnitude greater than those for nickel [24 - 26].

A major drawback to the use of Raney nickel in alkaline fuel cell assemblies is its pyrophoric nature [27], which can lead to spontaneous ignition in oxygen containing atmospheres and to allow their use as an electrode material the Raney nickel catalyst is commonly stored in distilled water where its pyrophoric nature is controlled. Nevertheless, in order to locate the catalyst onto the supporting electrode structure a passivation process is carried out that typically entails the use of hydrogen peroxide leaching resulting in a thin layer modification of the surface. This process creates thin oxide layers which trap entrained hydrogen in the surface pores. Once the catalyst is in place on the anode, the passivation step is reversed and the material is reactivated by heating the electrode in a hydrogen atmosphere [28].

fuel cells applications has been the unsatisfactory performance, particularly at higher electrical currents, and stabilities far below the 4000 hours which could typically be obtained from a platinum electrode, [24]. However, as can be seen in Fig. 1, electrodes manufactured by a gas atomization process demonstrate operational fuel cell voltages (OCV) that are comparable to that of a platinum electrode, for example, 440 mA cm^{-2} (Doped-Gas Atomised, Doped-GA) *cf.* 500 mA cm^{-2} (Pt) at a cell voltage of 750 mV. It is also worth noting that sponge nickel produced by gas atomization but without doping also offers increased levels of activity when compared to the more traditional methods of manufacture: 310 mA cm^{-2} (Gas Atomised, GA) *cf.* 190 mA cm^{-2} (Cast and Crush, CC) also at 750 mV.

Approximately 2.5% of primary nickel metal (a majority of which is in the form of sponge nickel) is used for catalytic purposes, which in 2001 - according to Larsen and Tyle [29] - was equivalent to ~2144 tonnes in Europe. Traditionally sponge nickel has been produced by the Cast and Crush (CC) method, which involves the casting of a mixed nickel-aluminium ingot (normally 50:50 wt. %). This ingot is then subject to a crushing process that produces a 50-100 μm powder with the characteristic appearance shown in Fig.2a.

An alternative method for the production of Ni-Al powder is to use the Gas Atomization (GA) method. GA has a number of advantages when compared to the Cast and Crush (CC) method, in particular the ability to produce smooth and spherical particles with extremely fine metallurgical microstructures that are much more refined than those produced by CC (Fig. 2b) with diameters ranging from $>5 \mu\text{m}$ to $\sim 45 \mu\text{m}$. The fine and spherical nature of the powder results from GA process where a high velocity gas flow is used to break up a stream of molten Ni-Al alloy emerging from a nozzle into a fine spray of liquid metal droplets,

resulting powder – whether produced by CC or GA – is then subsequently leached with sodium hydroxide (caustic leaching) to produce the required nanoporous nickel structure by the removal of the aluminium alloy component. Powder size distributions (PSD) for leached powders were, for the gas atomisation D_{10} –23.5 μm , D_{50} –34.8 μm and D_{90} –46.9 μm , and for the cast and crush, the PSD were D_{10} –10.7 μm , D_{50} –37.2 μm and D_{90} –72.4 μm .

Overall using the GA method to produce powder has distinct advantages when compared to CC:

- Ingot crushing is avoided.
- Particle size and distribution can be more tightly controlled reducing the need for intensive post-processing to recover required particle sizes.
- Individual particle cooling rates can be controlled resulting in enhanced microstructures that subsequently - post-leaching - have higher surfaces areas, which increase catalytic activity.

Depending on the cost and required functionality sponge-type catalysts can also be made from other metals like platinum and palladium or copper and cobalt also using the Raney process [30, 31]. In terms of functionality catalyst activity, selectivity, lifetime and surface area for the required reaction are the main considerations. The level of activity of the catalyst is related to the amounts of hydrogen that can be absorbed on the catalyst's micro-surfaces and which can reach levels between 25–100 ml of hydrogen per gram of nickel resulting in enhanced reaction. The more readily the hydrogen can be absorbed, the greater the resulting activity, though this level of catalytic activity reduces over time as a result of reactive site

poisoning and loss of catalyst integrity. Catalyst lifetime is also heavily influenced by the operational conditions of the fuel cell like temperature/pressure and cleanliness of the fuel source.

The aim of this paper is to carry out a comparative Life Cycle Assessment (LCA) of new gas atomized sponge nickel catalysts and evaluate their performance as electrodes against both cast and crush sponge nickel and the platinum standard currently used in commercial alkaline fuel cell applications. The purpose is to determine whether the increases in catalytic activity/performance observed on the laboratory scale could lead to any significant environmental improvements over the standard platinum electrode; with both un-doped and doped catalysts (where elements like Fe, Mo, Sn or Ti are added to further enhance catalytic activity) considered as part of this assessment.

2. Life Cycle Methodology

Generally following ISO14040 guidelines, the objective of the LCA was defined as “to understand the environmental impact of the standard sponge-nickel catalyst over its lifetime as an electrode within an alkaline fuel cell and whether refinement of the standard catalyst could lower the overall impact”. The scope in this application of sponge nickel was limited to the effects of materials substitution of a single component within the fuel cell, specifically, the contribution made by the catalyst within the electrode, as opposed to the entire fuel cell. To this extent, it was decided to ignore additional materials used in its construction (e.g. conductive carbon or polymer binders) as similar quantities were used whether the catalyst was sponge nickel or platinum catalysts. Furthermore, as the electrodes are interchangeable regardless of the catalyst used, the overall materials used in the fuel cell (e.g. housing and

assessment.

The compilation of inventory data for each of the steps in the process was done with the objective of prioritising process data which had itself been derived to ISO14040 standards. The benchmark standard catalyst chosen for comparison was one produced via the CC route, as opposed to the novel catalysts which were produced via the gas atomization route. The LCA was carried out from the perspective of the catalyst alone, i.e. it did not include the materials or life of the reactor itself and from a temporal perspective was based on the number of batches the catalyst would survive in a particular reactor.

In LCA methodology, there are global initiatives organised by the United Nations Environment Program (UNEP) and the Society for Environmental Toxicology and Chemistry (SETAC) which have resulted in the definition of relative weighting factors of emissions and their normalised contribution to environmental areas of impact such as the Global Warming Potential and Acidification Potential. These agreed values result in standardisation of practises and methodologies nominated according to the date and origin of the convention, such as CML2001 (Institute of Environmental Sciences, Leiden, Netherlands 2001), Eco-indicator99 and EDIP97 (Danish Environmental Design of Industrial Products, 1997), for which more details can be found in Dreyer *et al.* [34]. The first two are so-called mid-point approaches, and the latter is an end-point approach. In this work we have primarily used the CML2001 approach (based on a 100 year timescale for assessing any effects), which is accepted as having a robust account of impact factors in the Climate Change impact category and is less susceptible to the differences between methodologies as are impact categories like Human Toxicity and Ecotoxicity.

Where the current LCA work departs from standard guidelines is due to the fact that key data in the lifecycle is predictive, as the catalysts being examined are still at the laboratory/pilot scale as opposed to mass production, and there has not been to date a complete LCA done by the catalyst producers for this particular catalyst. Furthermore, a high level of uncertainty for the data of some processes was unavoidable, and estimating parameters were very difficult to find in public domain sources particularly in the operational and recycling stages of the lifecycle. Another departure from the ISO14040 guidelines is that it has not been possible in this study to implement due process involving independent verification (via third parties and life-cycle inventory committees) of data measurements made at process level of production, waste, energy and emissions. In addition, argon is used in significant quantities (0.33 kg per kg NiAl) throughout the process at various points and from various sources but the impact has been simplified to a power input requirement equivalent to 0.13 kWh/kg [35].

Nevertheless, the results of this work should still be of significant interest to what is a large industrial sector.

3. Life Cycle Assessment

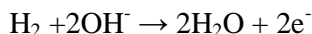
Sponge nickel catalyst life cycle phases, including the main process stages and key data requirements considered in the LCA are shown in Fig.3 and were implemented in GaBi 4.3 (Software by PE International for lifecycle assessment) [36]. As outlined, all the stages require energy in numerous forms and from different sources, with each stage resulting in the production of emissions. In order to determine the inventory data and all primary material/consumable process information has been compiled from as wide a range of sources as possible and calculated Gross Energy Requirements (GER), are also scrutinised with referenced to the available literature as outlined in the work of Eaves [37].

3.1 System Boundaries

Emissions were based on the assumption of global outputs and pre-existing ISO14040 life-cycle inventory data were utilised where available like nickel [38]. It is understandable that a certain amount of deviation may occur when extrapolating global averages from European-based production data, as highlighted by both the Nickel [38] and Aluminium Institutes' [39] in their life-cycle assessments, and potentially the scale of these discrepancies could be substantial. Such deviations can be understood, for example, by considering aluminium production. The electrolysis process used in aluminium production is responsible for approximately 85% of the total energy requirement of manufacture and as such the difference in levels of emissions are large as result of whether the electricity source is a fossil fuelled power station or hydroelectric power plant. As a consequence the utilisation of available data should be approached with caution as it often does not include information from countries like India, Russia and China - that have very different mixes of electrical power generation when compared to somewhere like Europe - often due a lack of comprehensive data. Moreover, additional information for catalyst production in the form of the supply chain and materials transport is often omitted, which could result in an additional 10-20% more Global Warming Potential (GWP) emissions.

3.1.1 Alkaline Fuel Cell Electrode

An alkaline fuel cell (AFC) generates electricity by converting of hydrogen and oxygen chemical energy into electrical current by the hydrogen oxidation reaction:



(1)

in the region of approximately 80%) and have a thermodynamic fuel cell voltage of 1.23V at 25°C [40]. Although low temperature (25 to 75°C) AFC's have preferentially used platinum and platinum group metals either separately or in combination, the use of Raney-nickel catalyst has been subject to increasing interest due to it being one of the most active non-noble metal catalysts for the hydrogen oxidation reaction [41, 42].

The fuel cell chosen for this study was the HC-100 AFC of Oy Hydrocell, Finland (Fig. 4), which can use either a platinum/palladium or a sponge nickel electrode interchangeably, where all other parts, heat exchangers, casing etc. remain exactly the same. Thus, in this LCA, production of other materials such as plastics, steels and copper alloys, which are required in the manufacture of this product, were deemed outside the system boundary. However, the activity of the catalyst is directly related to the hydrogen consumption, thus details of the production method of hydrogen are critical in this application. To this extent, three methods were looked at, namely steam reforming, hydrocarbon cracking and electrolysis.

3.1.2 Function and Functional Unit

In the case of the LCA outlined here, the function of the sponge nickel is defined to be the production of 20Wh of electricity from the hydrogen fuel cell. The overall functional unit of the LCA has been chosen to be the weight of catalyst required in a single electrode within the alkaline fuel cell set-up.

3.2.1 Upstream Materials

The extraction routes and metallurgical processes used to extract the primary metals like platinum and the aluminium/nickel used in sponge nickel are not within the scope of this paper and hence a full list of the inventory data is not included although some of this data is readily available [43 - 45]. Instead Table 1 is included as a summary of the overall Gross Energy Requirements (GER) in MJ/kg of product and an average of quoted greenhouse gases (GHG) in kg CO₂-equivalents per kg of product and follows previous estimations detailed by Lavery *et al.* [46].

Platinum Platinum was the only metal required in this analysis for which LCA data was not readily available as ISO measured data – a difficulty also noted by Staffell and Ingram [16]. The Anglo-Platinum mining operation in South Africa was modelled from data by a paper by Jacobs [47], however, the energy calculated here includes that used to obtain co-products (Ni, Cu, Os, Pd) and no allocation was used for these by-products, so this energy is expected to be on the high side.

Nickel Primary nickel data was taken from the Nickel Institute LCI report [38], with the production of >99%Ni (Class 1) nickel standard chosen as the benchmark in contrast to nickel oxide or ferronickel manufacture. The LCA data, from 2000, contains information on approximately 56 to 64% of the total annual world production of nickel, but excludes any specific details of Chinese or Russian production. As nickel processing routes can vary depending on the type of ore used (whether sulfidic or oxidic) the data is based on an average of both pyrometallurgical and hydrometallurgical extraction routes.

Aluminium LCA report [39]. Bauxite mining data represents information from approximately 49% of worldwide operations (10 mines), data for alumina refining from 59% of world-wide operations (24 refineries) and electrolytic aluminium production information is based on 55% of world-wide operations (75 electrolysis plants), but the report excludes any Chinese production data. In this work, the Bayer process for alumina production, followed by Hall-Heroult electrolysis for the alumina reduction to aluminium and finally casting of the aluminium into an ingot was chosen as the representative aluminium production process.

Dopants (Mo, Ti, Sn, Fe) In order to boost catalytic efficiency dopants can be added in the order of 1-3 wt.% and usually comprise of tin and transition metals like molybdenum, titanium and iron. Information for tin (Sn) was taken from data that was part of the existing GaBi Lean Database, and cross referenced with the Tin Institute's (ITRI) life-cycle inventory data [48]. Landfield Grieg's [49] LCI report for the International Molybdenum Association (IMoA) only contains data on the production of technical grade molybdenum trioxide (MoO_3), so additional processes in the form of a *sublimation* phase to obtain pure MoO_3 and a *reduction* stage to obtain pure Mo powder were required. Implemented data for titanium production via the Kroll process was obtained from Norgate *et al.* [50, 51]. A representative data inventory for iron was also assembled based on data from Norgate *et al.* [52] for the production of pig iron in a blast furnace. The inclusion of dopants is for the sake of completeness as they only contribute a relatively small environmental GWP burden, their presence does however have a significant impact in terms of recycling and disposal as outlined in more detail below.

hydroxide (caustic soda) and industrial water (both processed and pure) was obtained from information readily available in literature and from the GaBi Lean Database [53, 54]. The exception to this was Argon, which is consumed in significant amounts during the gas atomization process and for which there was no data. Nevertheless, according to the literature [35, 55, 56], a value of 0.13 kWh/kg is required to produce argon by cryogenic fractional distillation, resulting in a 7% increase in embodied GHG emissions over the standard cast and crush Ra-Ni.

3.3 Manufacturing

3.3.1. Gas Atomized (GA) Powder Production Route

Table 2 presents inventory data extracted from a number of papers available in the literature [55, 57 - 59]. The gas atomizers used in this study were assumed to feature induction melting methods, which offer efficiency in the region of 50% and this was subsequently used to determine an estimate for the practical melting energy. In addition, out-of-range particles produced by gas atomization can be recycled via collection and re-melting in the following run which results in an overall process efficiency of ~95%. The theoretical energy requirements for melting were calculated by using thermodynamic properties including latent heat and specific heat capacity. Overall, these melting energy requirement values were compared to those detailed in the work of Dunkley and Aderhold [60] and they showed a good correlation. The levels of alloy loss were assumed to be insignificant ($< 0.1\%$) as the whole melting process is performed under an argon atmosphere and any material that is lost due to volatilisation is collected by cyclone filters, which can be later recycled. All measurements detailed in this paper were performed on a small pilot scale gas atomizer without the addition of an expensive gas recycling component. This means that the system utilised was unable to recycle the argon gas, which would have increased the efficiency of the

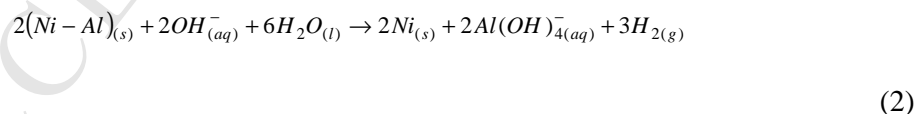
system could reduce argon use to < 0.007 kg/kg Ni-Al alloy.

3.3.2 Cast and Crush (CC) Powder Production Route

As the name suggests the production route comprises of two distinct steps, melting (casting) and crushing (i.e. milling/grinding/attrition). As with the GA process, melting was assumed to occur under an argon blanket with a 50% efficiency and known NiAl thermodynamic properties allowed energies to be estimated at between 0.60-0.8 kWh/kg. From the work of Pourghahramani [58], the grinding/crushing step - based on the average energy required for the mechanical alloying of all materials to different sized particles - was deemed to be 0.044 kWh/kg. This value, however, may be an overestimation as Ni-Al material is brittle in nature and as such needs lower than average energies of attrition.

3.3.3 Caustic Leaching

Caustic leaching is a batch process that is used for Ni-Al powders produced by both CC and GA, which uses a 20 to 30 wt. % aqueous sodium hydroxide (NaOH) solution to almost totally remove the aluminium to leave sponge nickel and a sodium aluminate (NaAlO₂) by-product as described by Devred *et al* [59, 61] and outlined in Eq. (2):



This procedure can be used to process up to a tonne of material at once and this leaching removes almost all of the aluminium from the powder leaving a porous structure with a very high surface area, which is linked to the catalytic activity of the powder [62]. The sodium aluminate solution can be removed from the solution and is typically sold for use in paper manufacturing or water treatment. Recovery of aluminium from the solution is also possible

the reaction of sodium hydroxide with the Ni-Al powder is highly exothermic which allows the desirable process temperature of 80°C for the solution to be maintained by the controlled addition of NaOH and the process of leaching takes about 3 hours to complete. Once leaching is complete, the sponge nickel can then be rinsed and stored in distilled water until required. In reality, higher concentrations of NaOH solution are used in an effort to reduce precipitation of aluminium hydroxide ($\text{Al}(\text{OH})_3$) that can cause pore blockage in the sponge nickel and thus affect ensuing catalytic functionality.

3.3.4 Alkaline Fuel Cell Electrodes

The typical platinum loading of the electrode in the Hydrocell Oy AFC fuel cell is 0.4 g compared to that for 1.5 g of sponge nickel loading, as measured by Eaves [37]. The activity of the different catalysts can be directly related to the consumption of hydrogen and in the current work, a constant power output of 20W was used as the benchmark for the application of the fuel cell [37]. The respective hydrogen consumption for this power requirement for the platinum electrode, the gas atomized sponge-Ni electrode and the cast and crush sponge-Ni electrode, respectively is shown in Fig. 5.

Three electrode types were considered: a Platinum electrode (PTE), a sponge nickel electrode produced by gas atomization (Ra-Ni-GA) and a sponge nickel electrode produced by the cast and crush process (Ra-Ni-CC) with a typical electrode lifetime defined as up to 3900 hours. In terms of hydrogen (H_2) requirements for the lifetime of the electrode, the activity differences of the catalyst lead to a difference in lifetime consumptions of hydrogen, based on the measured consumptions. Thus, over the lifetime of 3900 hours of the fuel cell, the PTE required of 3.2 kg H_2 ; the Ra-Ni-GA electrode required 3.91 kg H_2 and the Ra-Ni-CC

production routes were considered for the life cycle assessment, namely steam reformation (89.22 MJ/kg), hydrocarbon cracking (71.84 MJ/kg) and electrolysis (16.07 MJ/kg), all of which are readily available as inventory data from the European Plastics Association and are provided within the GaBi software [63-65], and for which up-to-date production energy and emissions are documented in an LCA by Cetinkaya et al [66]. The GER and GWP values used in this analysis are given in Table 1. The values outlined are average (Gross) calorific energy requirements for the production of 1kg hydrogen at standard pressure, and account for average efficiencies which can be obtained by the various processes. It should be noted however that net calorific energy requirements from the electrical input will vary depending on the local source of electricity mix.

It has been previously found by Jarvis and Voss [67] that gas atomized powder sponge nickel has enhanced activity when compared to that produced by the standard CC method. This increased catalytic activity leads to reductions in hydrogen fuel consumption and a reciprocal increase power output (as seen in Fig. 5). These changes in relative activity were used as a basis to approximate the levels of hydrogen fuel reduction possible with sponge nickel (with and without the addition of a dopant), when compared to both the standard industrial platinum catalyst and CC sponge nickel. However it needs to be stressed that such estimates are based on limited literature sources that need further validation. Over time the catalyst material becomes contaminated and degrades, which leads to a reduction in performance and a subsequent replacement of the catalyst. There are a number of reasons that affect catalyst lifetime like operating temperature and pressure within the fuel cell, purity of the hydrogen gas source and presence of competing hydrogenation reactions.

3.4.1 Regeneration of Spent Catalysts

In 2002 non-precious catalyst use in Germany alone was approximately to 9000 t/year [68], of which, only a tiny proportion was being recycled. This was primarily due to the expensive recycling costs when compared to the cheaper alternative of waste treatment (to prevent the leaching of heavy metals into the soil followed by landfill. In addition work by Marafi and Stanislaus [69] estimated that for a Hydrotreating process the levels of have spent catalyst doubled over a decade (1996 to 2007) from 87,500 to 170,000 t/year.

Over the last decade a combination of high metal prices, increased landfill costs (primarily due to increasing regulations related to hazardous material disposal), increased spent catalyst availability and improvements in recovery methods means that spent catalysts now have the status of useful commodity with a positive net value, especially those with vanadium and molybdenum constituents. It is now common for catalyst producers to offer catalyst life cycle management where the catalyst is regenerated “ex-situ” by a specialist company

Spent catalyst recycling can be achieved by using hydrometallurgical or pyrometallurgical methods with the attained levels of regenerated catalytic activity being as high as 70 to 80% and a cost of 50% when compared to fresh catalyst. These regenerated catalysts are generally used in combination with fresh catalyst in ratios up to 50% and in terms of sponge nickel the regeneration recovery rate is approximately 90% with the associated energy consumption about 10 MJ/kg *cf.* 200 MJ/kg for primary Ni production [70].

The amount of spent sponge nickel catalyst that is dumped in landfill by European manufacturers is unknown as such disposal is expensive due to the toxicity to humans of metals like chromium, nickel and molybdenum highlighted by McLaughlin [71]. In the absence of such verifiable yearly data the inclusion of heavy metal water contamination from disposal of spent catalyst in landfill has not been possible.

3.5 Inventory Data Analysis

It is essential to highlight the dissimilitude between data quality utilised in the context of the LCA outlined here. For the two main constituent metals (nickel and aluminium) used in the catalyst manufacture there is a wealth of high quality data available from both the Nickel [38] and Aluminium Institute's [39], full ISO14040 LCA of the respective elements. For consumable materials used there is a distinct variation in standards varying from the good for water, sodium hydroxide and hydrogen (which are all part of the GaBi lean database) to low quality for argon where only the energy of production is considered [35, 55]. A similar situation is true for the doping materials where there is a broad range of standards from good as in the case of molybdenum (IoMA LCA, [49] to intermediate (tin data from the GaBi lean database) to poor for iron and titanium, where the LCA data is assembled from sub-models based on various sources. However, as the process has the most emphasis on the primary metals –nickel and aluminium – rather than consumables doping materials it can be considered that the overall quality of the upstream data is relatively good and is a suitable basis for the comparison of the standard industrial catalyst and the gas atomized sponge nickel.

materials consumption measured by partners in the LCA project and thus it can be regarded as being of an acceptable quality. This data does, nonetheless, have a bias towards the use of electrical energy and thus is more suitable for “carbon footprint” analysis, which – as previously discussed by Laurent *et al.* [72] - is a poor indicator of other environmental impacts.

The recycling and disposal phase contains the lowest quality of inventory data and it is here that a very high (99%) recovery rate is assumed, which is broken down into 50% recovery for catalytic applications; 49% downgraded; 1% lost/disposed of. As the emphasis of the LCA is catalyst comparison, the 1% loss is deemed to be an insignificant change when comparing one catalyst with another, thus would not affect the final outcome and understanding.

4. Results and Discussion

To achieve a similar level of functionality, the electrode for the alkaline fuel cell requires different weights (loadings) of platinum and Raney nickel catalyst of 0.4g and 1.5g, respectively [37]. The required loading of the sponge nickel catalyst was much higher 50-100 mg/cm² than the platinum loading of 2-5 mg/cm². By using data from the raw materials and manufacturing process it is possible to calculate the total energy of production for the different electrode materials and the results of which are shown in Fig.6. As can be seen, for the materials investigated, platinum mining and electrode fabrication has by far the largest demand for energy during the whole extraction and production process requiring an estimated total calorific energy of 9.3 MJ when compared to 1.25 MJ for GA sponge nickel and 0.45 MJ for CC produced sponge nickel. As can be expected these differing levels energy consumption have differing amounts of associated Global Warming Potential (GWP) varying

as shown in Fig. 6.

An overview of the results for the entire lifecycle energies (Gross Energy Requirement, GER) and emissions (Global Warming Potential, GWP and Acidification Potential, AP), are summarised in Tables 3, 4, and 5, allowing upstream production energies to be compared to the “in-service” energy usage which is in proportion to the energy used by the hydrogen production process used over the lifetime of the fuel cell. The hydrogen production processes considered are steam reformation, electrolytic and thermal cracking, as previously outlined by Cetinkaya *et al.* [66]. The overall consumption of hydrogen over a 3900 hour time-span is based on measured hydrogen consumption as outlined in Fig. 5. Also, indication of the catalytic mass activity was calculated from the experimental data which varied from 100 A/g for the platinum catalyst to 24 A/g for the doped Ra-Ni catalyst and to 10 A/g for the standard CC Ra-Ni catalyst. Highlighted in bold in each table are emissions from the gas-atomised and doped catalyst which invariably (regardless of hydrogen source) has the lowest environmental impact.

What is clear from the overview tables is that the upstream extraction and manufacturing emissions of the platinum is a considerable lifetime contributor to overall emissions. While the lifetime emissions of the fuel cell are, and should be dominated by the use of consumables, mainly hydrogen, it is clear that as the production of hydrogen moves towards a more sustainable sources (electrolytic-renewable energy), as opposed to fossil fuel (steam reformation – gas source), so that the production of hydrogen has minimal GHG emissions, so the upstream back-pack of emissions becomes most important. Platinum electrodes have an order of magnitude greater emission than nickel catalysts in production, and this would

required from the perspective of quantifying the long term recycling and re-useage issues, but this was not covered in the current lab-scale work and is an area best undertaken with better knowledge of pilot or larger-scale production volumes.

Fig. 7 and Fig. 8 show summary results from the LCA implementation in GaBi, where the inventory data described in the overview tables has been scaled to a central European output scale as determined by the CML2001 LCA methodology. *What is not so clear from the presentation of the results is that rarity and low percentile occurrence of platinum in the earth results in its high price and it also being a contributor to abiotic depletion categories of the life cycle analysis.*

The overall lifetime (accounts for both primary production and operational phases) reduction in GER energy, GWP and AP emissions is summarised in Table 6. This shows that for a 3900 hour lifetime of the electrode, there can be between 13-43% reduction in overall GER energy usage, between 40-74% reduction in GHG emissions, and a reduction of 64-85% in AP.

Whether one looks at the tabulated data (Tables 3 - 6), or the figures (Fig. 7 and 8), common results can be summarised as:

- Relatively small differences in activity of the catalyst can lead to large savings in hydrogen and energy use over the lifetime of the electrode.
- Although the gas atomisation route for manufacture of Raney-nickel requires more energy input than the industry standard cast and crush process, the overall production energy and emissions for an equivalent mass of Ra-Ni catalyst in the electrode are an order of magnitude lower than those required for a platinum-based electrode.

the activity and stability of the catalyst, at relatively little upstream energy and emission costs. In the electrode, the best gas atomised doped catalyst can give equivalent power outputs to a platinum based one.

- Even though platinum tends to have a higher mass catalytic activity than Raney nickel, longer lifetime payback times are required to overcome the upstream extraction and production, from the perspective of energy and overall emissions.
- This is particularly the case if hydrogen is produced via electrolysis using electricity from low carbon energy sources, as opposed to a fossil fuel-based production route such as steam reforming. In the former case, the emissions required in primary and manufacturing of the platinum (9 kg CO₂-eq.) are 3 times greater than the lifetime emissions associated with the production of the hydrogen itself (3.1 kg CO₂-eq.).

5. Conclusions

From the results of the comparative LCA it can be concluded that although the production of 1kg of sponge nickel catalyst by a Gas Atomization (GA) route requires only 13.5% of the energy when compared to that of the production of 1 kg of Platinum (but twice as much as standard Cast and Crush (CC) sponge nickel), doped GA sponge nickel offers a comparable catalytic activity. It is this catalytic behaviour that - based on the estimates outlined in this paper – have the potential to offer a reduction of the overall energy consumption, and a significant decrease in GHG emissions and hence an associated decrease in the Global Warming Potential (GWP) of the fuel cell over the complete lifetime of the sponge nickel catalyst when compared to the current industry standard.

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Fig. 1 Comparison of the Operation Fuel Cell Voltages (OCV) achievable for the different types of alkaline fuel cell (AFC) electrodes.

Fig. 2 SEM images of nickel-aluminide powder produced by (a) the Cast and Crush (CC) and (b) Gas Atomization (GA) methods.

Fig. 3 Overview of the LCA processes for the production and use of sponge nickel.

Fig. 4 (a) Schematic outlining the components of a typical alkaline fuel cell and (b) a picture of the fuel cell used for LCA.

Fig. 5 Amount of hydrogen gas consumption for a constant 20W/h output by (a) platinum electrode, (b) GA sponge-Ni electrode and (c) CC sponge-Ni electrode, respectively.

Fig. 6 Energy and Global Warming Potential (GWP) for the upstream production of materials and manufacturing of fuel cell electrodes.

Fig. 7 Overall energy requirements over the lifetime of alkaline fuel cells (AFC) with various electrodes and hydrogen production routes.

Fig. 8 Impact categories for all processes of alkaline fuel cell electrodes manufacture and operation. **Inset:** Expanded results for the AFC's using electrolysis derived hydrogen.

Table 1 – Material Gross Energy Requirements (GER) and Global Warming Potential (GWP).

Table 2 – Manufacturing process inventories.

Table 3 – Lifetime energy and emissions when hydrogen production route is by steam reformation.

Table 4 – Lifetime energy and emissions when hydrogen production route is by electrolysis.

Table 5 – Lifetime energy and emissions when hydrogen production route is by thermal cracking.

Table 6 – Overall reductions in emissions for different hydrogen manufacturing processes when comparing the total lifetime energy use and emissions for the best doped GA based catalyst compared to the platinum catalyst.

- Emissions during catalyst use greatly outweigh those of manufacture and recycling
- Gas Atomised spongy nickel catalysts can have a markedly lower environmental impact
- Doped Gas Atomised sponge nickel shows behaviour similar to a normal AFC Pt electrode

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Principal product	Method of Production	GER	GWP	AP	References
		(MJ/kg per kg product)	(kg CO ₂ -eq. per kg-product)	(kg SO ₂ -eq. per kg-product)	
Primary Metals					
Platinum (Pt)	Smelting/refining	102,213-179,779	24,600-43,303	104-183	[37],[47],[48]
Nickel (Ni)	Pressure leaching	114-239	11.4	0.130	[44, 45]
	Smelting/refining				
Aluminium (Al)	Bayer,	180-211	22.4	0.131	[43][51]
	Hall Heroult				[23-25]
Dopants					
Iron (Fe)	Blast Furnace /	22	2.3	0.02	[24]
	Pig Iron				
Tin (Sn)	Smelting/Refining	200	17	0.5	[48]
Molybdenum (Mo)	Roasting	175-203	11-13	0.32	[48]
Titanium (Ti)	Becher and Kroll	361	35.7	0.23	[49]
Gases					
Hydrogen (H ₂)	Reformed steam	89.22	4.20	0.07	[53, 63]
	Water electrolysis (Grid/Wind)	16.07	0.95	0.003	[53, 64]
	Thermo-Cracking	71-74	1.20	0.002	[53, 65]
Argon (Ar)	Cryogenic distillation/Air separation	0.32-0.48	0.0654	0.000157	[35, 55]
Other consumables					
Sodium hydroxide	Electrolysis (Chloralkali)	21	1.2	0.001	[54]

Table 2.

Process	Major inputs/outputs	Value	Functional Units	References
Gas Atomization	Melt Alloy: Ni-Al (+dopant Fe, Mo, Sn, Cr)	1	kg	Calculated
	Electrical energy to melt (+200 °C superheat)	0.7-0.8	kWh/kg Ni-Al	[57]
	Argon gas	0.333	kg/ kg Ni-Al	[55]
	100µm powder yield	95%	of Ni-Al alloy	
	Ni-Al losses (dry cyclone filter collection)	0.01%	of Ni-Al alloy	
	Output: 100µm Ni-Al powder	0.95	kg/kg Ni-Al	
Cast and Crush	Melting: Electrical energy (+200 °C superheat)	0.7-0.8	kWh/kg Ni-Al	Calculated
	Crushing: Electrical energy (1m-2cm)	0.001	kWh/kg Ni-Al	[58]
	Grinding: Electrical energy (2cm-200µm)	0.01	kWh/kg Ni-Al	[58]
	Milling: Electrical energy (200µm-10µm)	0.033	kWh/kg Ni-Al	[58]
	100µm powder yield	100%	of Ni-Al alloy	
	Ni-Al losses (filter collection)	0.01%	of Ni-Al alloy	Estimated
	Output: 100µm Ni-Al powder	0.99	kg/ kg Ni-Al	Calculated
Caustic Leaching	100µm Ni-Al powder	1	kg	
	NaOH (20-30wt.% Sodium Hydroxide)	3-4.5	kg/kg Ni-Al powder	[59]
	Leaching water (distilled)	15	l/kg Ni-Al powder	[59]
	Heating for 3 hours at 80°C (electrical energy)	0	kWh/kg Ni-Al powder	Exothermic
	Storage water (distilled)	50	l/kg Ni-Al powder	[59]
	Output: Sponge-Nickel (Dry)	0.5-0.3	kg/kg Ni-Al powder	Calculated

Catalyst material	Energy MJ / GHG kg CO ₂ -eq. / AP kg SO ₂ -eq.			A/g	Litres
	Primary and Manufacturing	Operational	Totals	Mass Activity	Total H ₂ use
Cast and Crush Ra-Ni	1.00 / 0.018 / 0.000	585.76 / 27.57 / 0.045	586.76 / 27.59 / 0.046	10.00	78,390
Gas atomised Ra-Ni undoped	1.27 / 0.037 / 0.001	317.65 / 14.95 / 0.025	318.93 / 14.99 / 0.025	13.33	42,510
Gas atomised Ra-Ni doped	1.28 / 0.041 / 0.001	288.51 / 13.58 / 0.022	289.79 / 13.62 / 0.023	23.33	38,610
Platinum	40.89 / 9.000 / 0.042	291.42 / 13.72 / 0.023	332.31 / 22.72 / 0.065	100	39,000

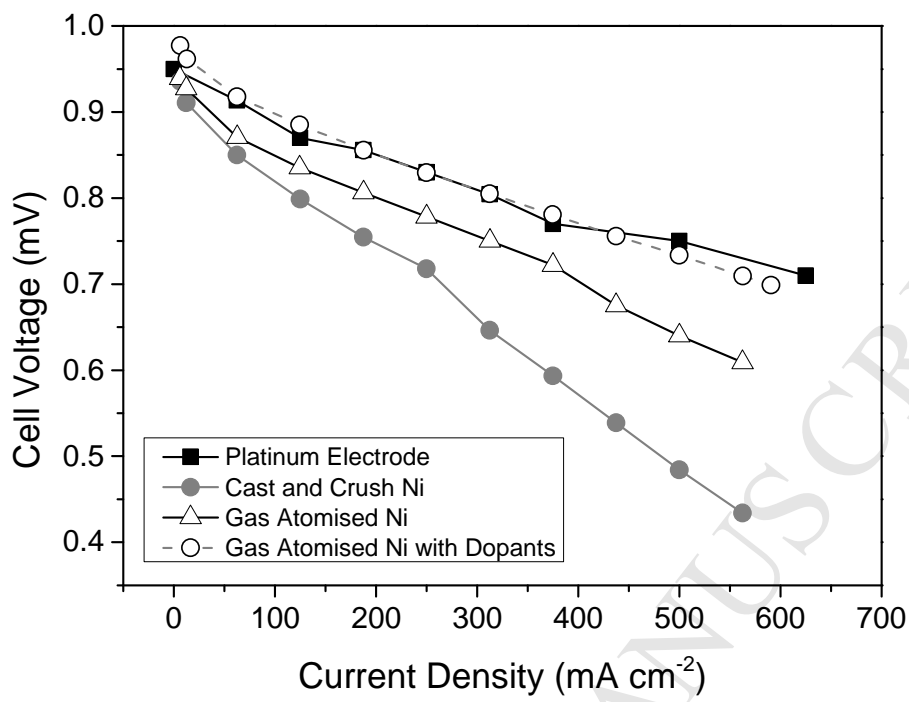
Catalyst material	Energy MJ / GHG kg CO ₂ -eq. / AP kg SO ₂ -eq.			A/g	Litres
	Primary and Manufacturing	Operational	Totals	Mass Activity	Total H ₂ use
Cast and Crush Ra-Ni	1.00 / 0.018 / 0.000	105.5 / 6.24 / 0.021	106.51 / 6.26 / 0.021	10.00	78,390
Gas atomised Ra-Ni undoped	1.27 / 0.037 / 0.001	57.21 / 3.38 / 0.011	58.49 / 3.42 / 0.012	13.33	42,510
Gas atomised Ra-Ni doped	1.28 / 0.041 / 0.001	51.97 / 3.07 / 0.01	53.25 / 3.61 / 0.011	23.33	38,610
Platinum	40.89 / 9.000 / 0.042	52.49 / 3.10 / 0.01	93.38 / 12.10 / 0.052	100	39,000

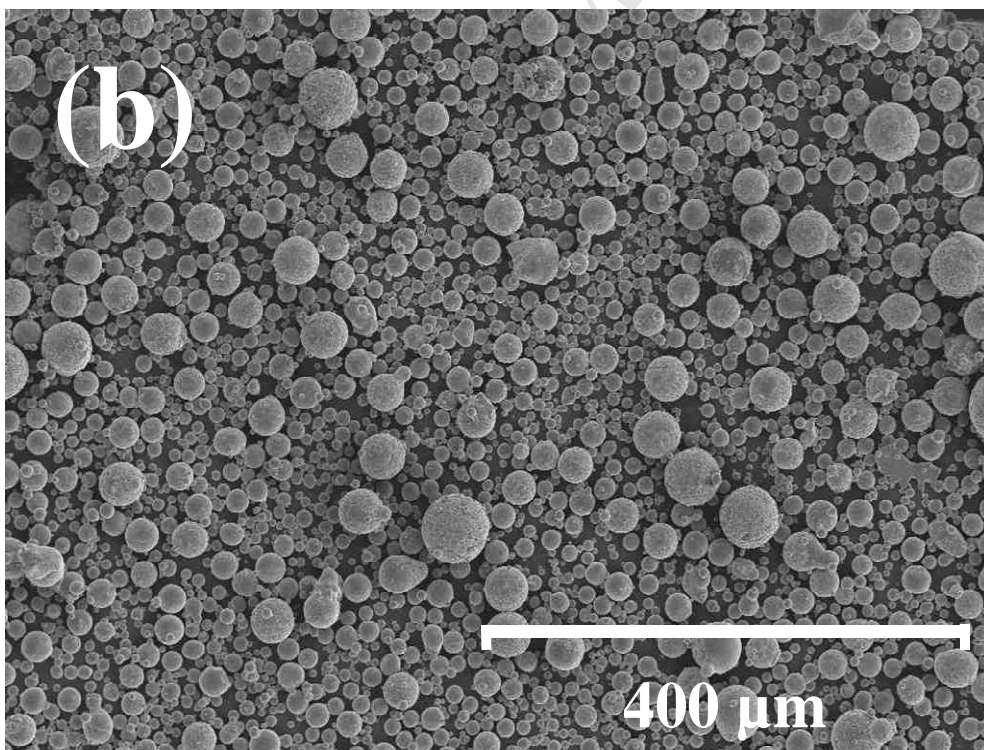
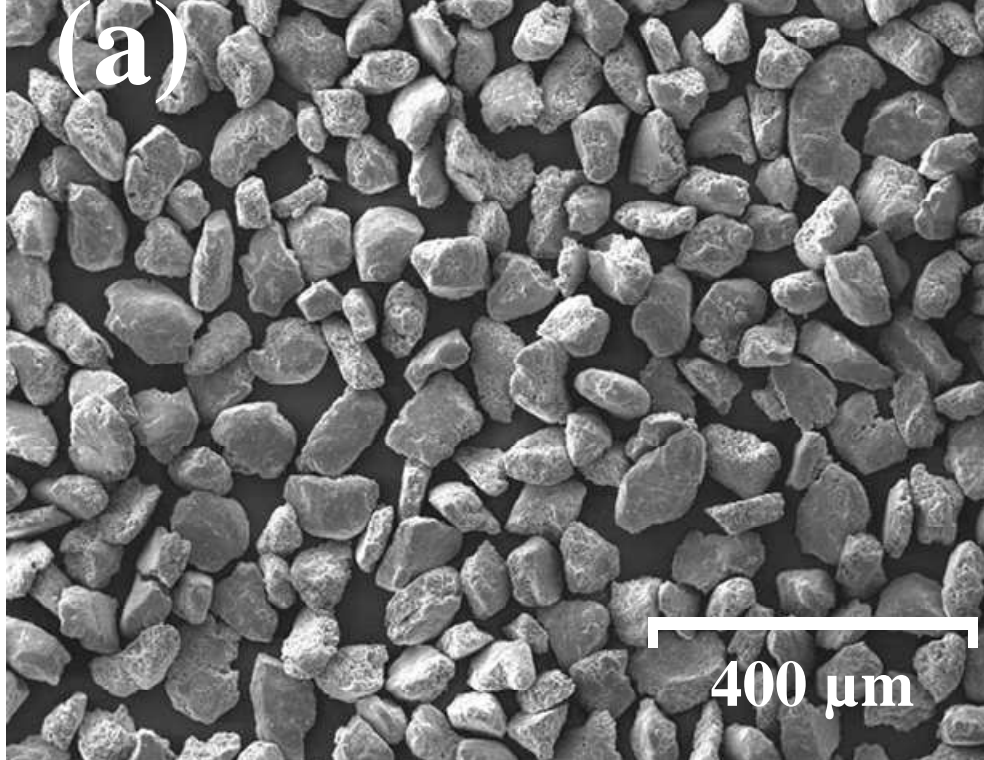
Catalyst material	Energy MJ / GHG kg CO ₂ -eq. / AP kg SO ₂ -eq.			A/g	Litres
	Primary and Manufacturing	Operational	Totals	Mass Activity	Total H ₂ use
Cast and Crush Ra-Ni	1.00 / 0.018 / 0.000	471.7 / 7.88 / 0.013	472.66 / 7.90 / 0.014	10.00	78,390
Gas atomised Ra-Ni undoped	1.27 / 0.037 / 0.001	255.7 / 4.27 / 0.007	257.05 / 4.31 / 0.008	13.33	42,510
Gas atomised Ra-Ni doped	1.28 / 0.041 / 0.001	232.3 / 3.88 / 0.006	233.59 / 3.92 / 0.007	23.33	38,610
Platinum	40.89 / 9.000 / 0.042	234.7 / 3.92 / 0.007	275.5 / 12.92 / 0.049	100	39,000

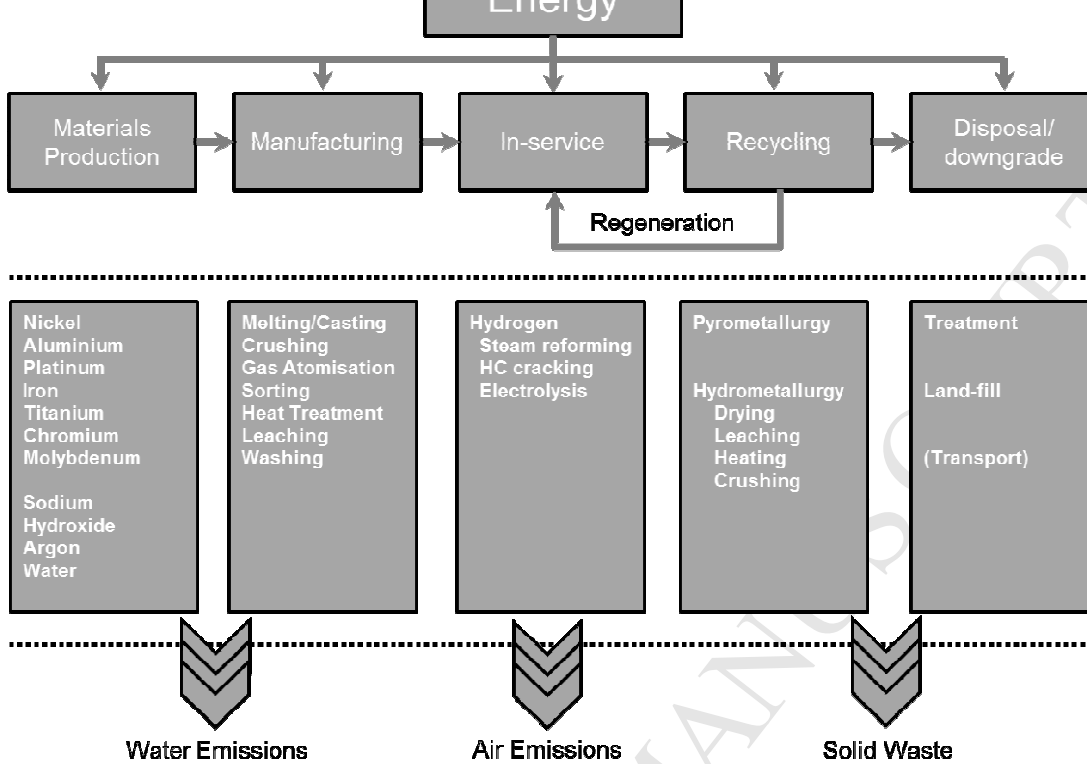
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Hydrogen manufacturing process	GER	GWP	AP
Hydrogen by steam reforming	13%	40%	64%
Hydrogen by thermal cracking	15%	70%	85%
Hydrogen by electrolysis	43%	74%	78%

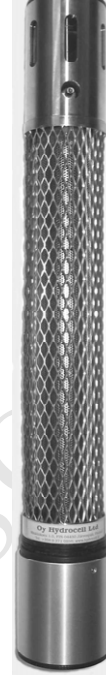
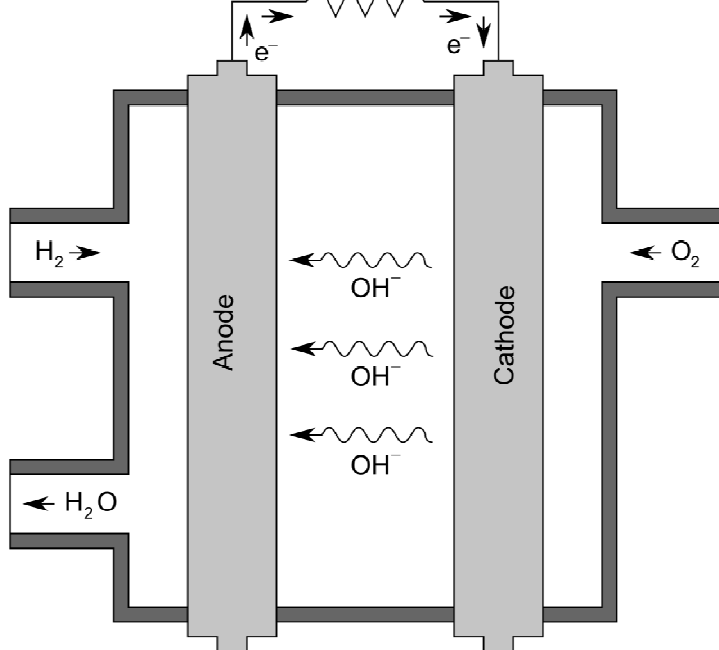
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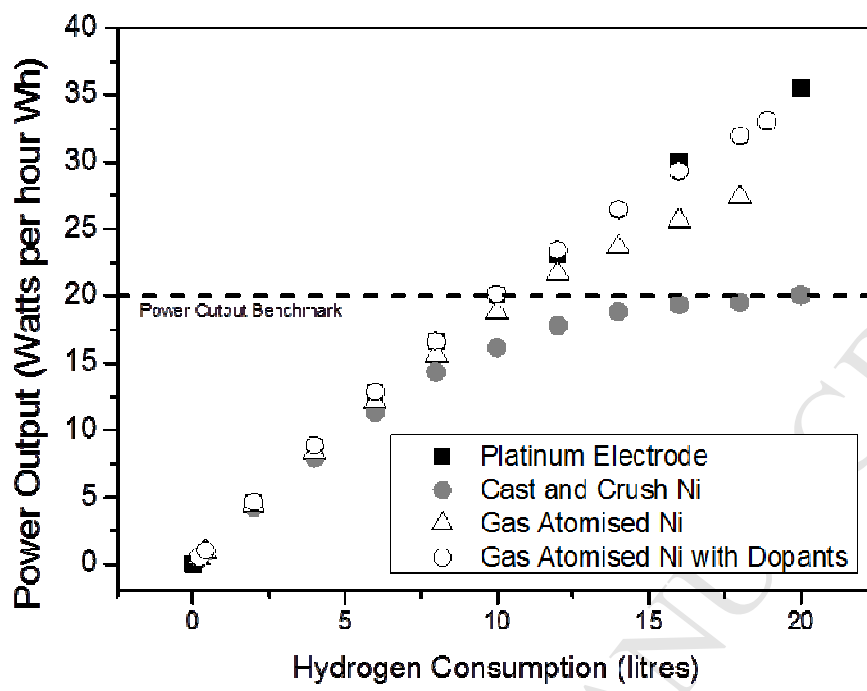


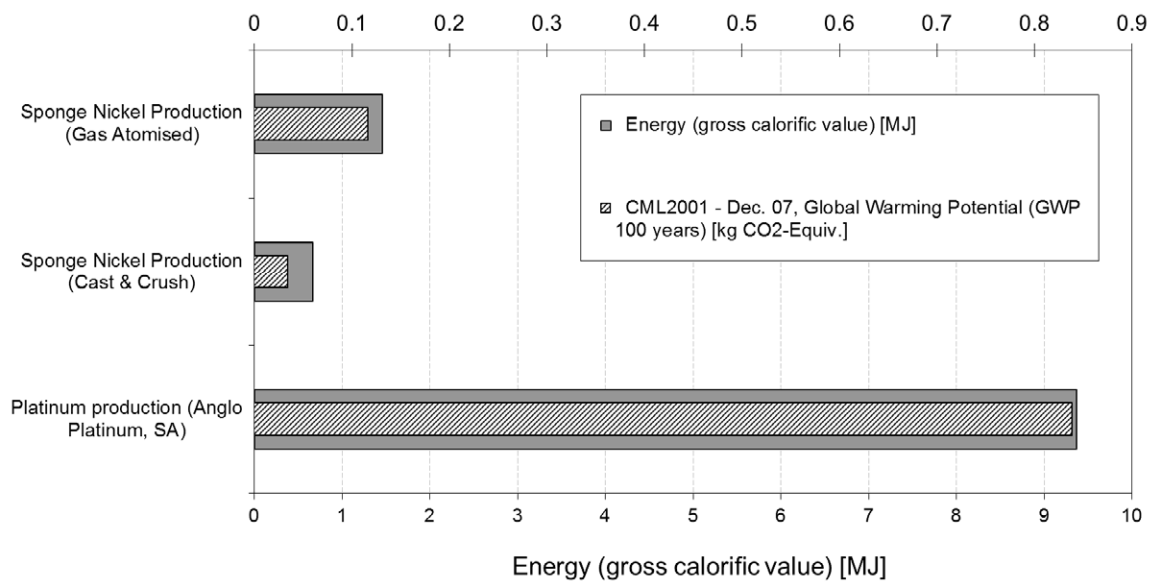


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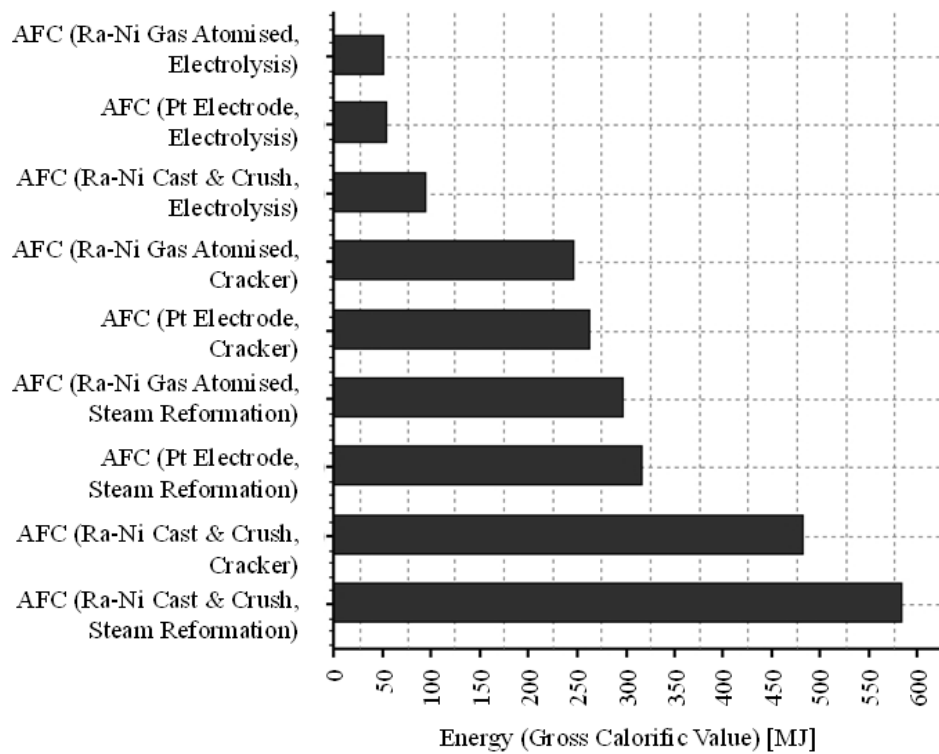


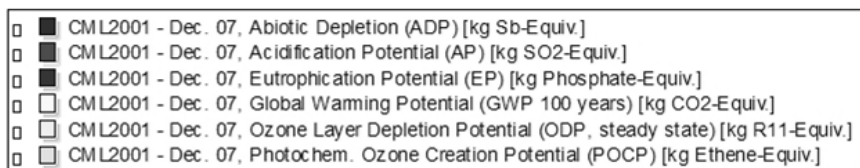
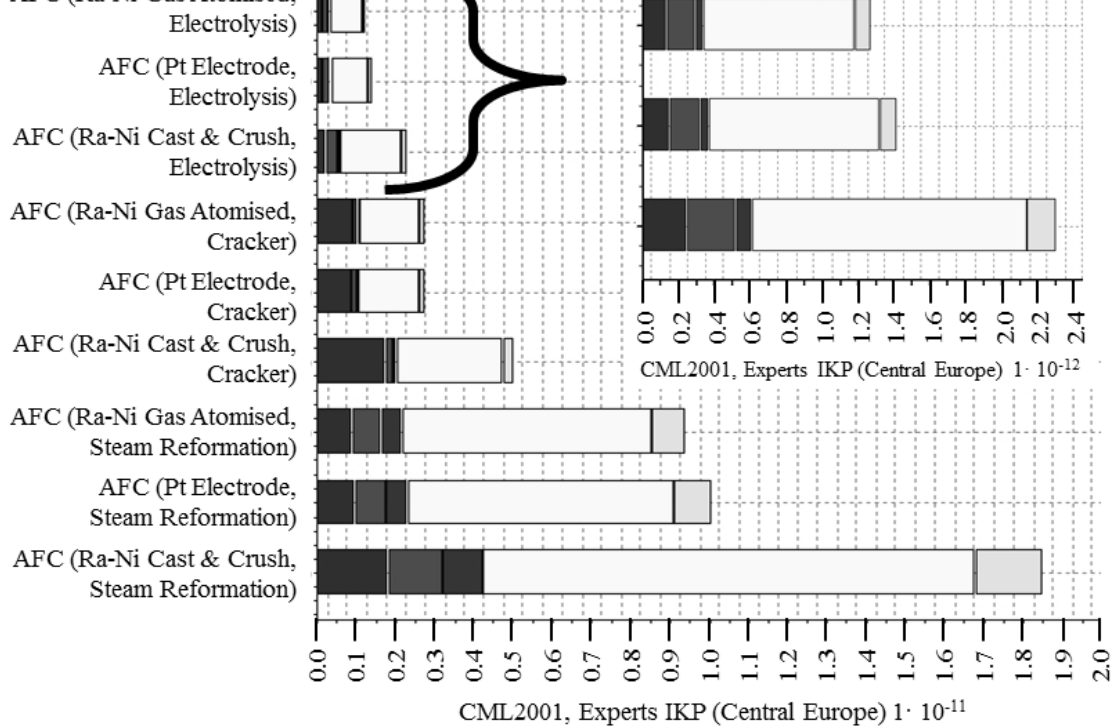
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