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Utilizing Life Cycle Assessment to support the environmentally friendly design of hydrogen generation from magnesium alloys: Offering a second life to waste

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Abstract

Hydrogen has emerged as a promising future energy source, but efficient, eco-friendly production and storage remain challenging. Life Cycle Assessment (LCA) is crucial for evaluating environmental impacts and guiding eco-design strategies. Among emerging techniques, magnesium hydrolysis for hydrogen production has gained attention. The presented LCA of a patented Mg-alloy with carbon and nickel additives identified nickel as the primary environmental impact contributor. To mitigate this, using ternary phases with minimal nickel content is proposed. Moreover, leveraging magnesium waste presents an opportunity to give alloys a second life, while enhancing the recycling of the resulting product, Mg(OH)₂. Although LCA results show that magnesium hydrolysis has a lower environmental impact than steam reforming, further industrial-scale research is needed. This highlights the importance of ongoing investigation and refinement of magnesium hydrolysis as a green energy solution.

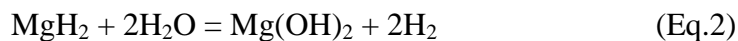
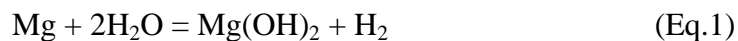
Keywords: hydrogen, magnesium waste, LCA, eco-design, metals recycling

1 Introduction

In recent years, hydrogen has emerged as a promising candidate for the future of energy. Despite the long-term interest, the environmentally benign production and the storage of hydrogen are still a challenge. Although there might be a mass production of hydrogen using renewable energy in the long term as well as the use of vegetable oils as an option for producing low carbon hydrogen [1,2], nowadays fossil fuels are the major source for its production [3,4]. Natural gas is a major source accounting for 47 %, fossil oil for 30 %, 19 % from coal, 3.9 % from electrolysis and the remaining 0.1 % from other processes [5]. Hydrogen can either be stored in the form of molecular hydrogen in pressurized vessels [6], liquefied hydrogen tanks [7] and carbon materials [8]. Storage in the form of atomic hydrogen in metal hydrides [9] or in the form of hydride ion in protide compounds such as alanates [10] and borohydrides [11] offer other alternatives. There are a number of factors that need to be considered when thinking about storage options including weight or volume, security and cost.

Hydrogen production based on hydrolysis reaction with various materials has been heavily studied for decades (the number of published papers has grown from about 100 in 1980 to more than 1500 in 2020, and it is still growing). This approach shows promise due to its energy-efficient nature (operating at room temperature) and its ability to yield delocalized and relatively pure hydrogen. Various materials, including complex hydrides [12], metals, intermetallics [13,14], and metal hydrides [15], have been extensively documented in literature.

The prospect of hydrogen generation through magnesium (Mg) hydrolysis has garnered considerable interest for several reasons: its widespread availability, low density, cost-effectiveness, and electrochemical reactivity [16]. Magnesium and magnesium hydride react with water as illustrated by the following equations (Equations 1 and 2, respectively):



The theoretical hydrogen yield stands at 8.2 wt.% for magnesium (Mg) and 15.2 wt.% for its hydride, excluding water in the calculation. However, the hydrolysis reaction faces limitations due to the formation of a passive hydroxide layer. Various strategies have been explored to

enhance the hydrolysis performance of Mg powder [17,18]. Studies have shown that hydrogen generation can be enhanced through the interaction of magnesium with organic acids [18], application of ultrasonic irradiation (especially notable in the case of MgH_2) [19], or through ball milling with different additives [16,20].

In a prior investigation [16], the beneficial impact of carbon additives such as graphite and carbon fibers on the hydrolysis reaction of MgH_2 and/or Mg has been illustrated. Additionally, the synergistic effect of graphite and transition metals, such as Ni, was examined. Moreover, the viability of onboard hydrogen production through the hydrolysis of MgH_2 to support proton-exchange membrane fuel cells (PEMFC) has been successfully demonstrated [21].

An increasing number of studies are focusing on employing magnesium alloys for hydrolysis reactions rather than pure magnesium [17-18, 22-23]. This approach not only lowers costs by utilizing non-recycled alloys but also provides a second life for discarded alloys. However, while this method may seem appealing at first glance, it requires deeper scrutiny and comparison with other methods in terms of performance, as demonstrated in several papers [17-18, 22-23]. Additionally, comprehensive assessments of its environmental impacts (*i.e.*, considering human health, ecosystem, resources and global warming) throughout the product life cycle are necessary.

Life Cycle Assessment studies (LCA) have extensively explored numerous hydrogen production routes [4,24–26]. However, most of these investigations have primarily focused on steam reforming and electrolysis, with variations in energy sources to evaluate their impacts. Concerning carbon dioxide emissions, studies indicate that water electrolysis powered by renewable energy sources is the most environmentally benign option [26]. To date, comprehensive life cycle assessment studies on hydrogen production from hydrolysis using Mg alloys are lacking. Only preliminary results have been outlined in a brief review concerning Mg-NdNiMg₁₅ composites [27]. Further in-depth analyses are warranted to gauge the environmental implications of this method.

The objective of this paper is to conduct a life cycle assessment (LCA) of magnesium hydrolysis for hydrogen production, examining both complete and partial processes, and to propose solutions to address the identified drawbacks. Specifically, we aim to demonstrate the application of LCA methodology in supporting environmentally friendly designs for hydrogen generation

from magnesium alloys, thereby giving a new purpose to magnesium waste. The LCA investigation focused on a patented Mg mixture designed for hydrogen generation, containing 5 wt.% of Ni and 5 wt.% of C [21]. The inclusion of nickel as an additive is crucial for enhancing galvanic coupling, particularly in scenarios involving magnesium waste utilization [8]. Additionally, the aluminum container of the Mg alloy was taken into consideration in our assessment.

2 Methods and materials

The experimental materials and methods are presented first in section 2.1, followed by the LCA methodology.

2.1 Experimental method

The powders used were prepared from Mg (Stream Chemicals, 99.9%), Ni (Stream Chemicals, 99.98%) and carbon (from Toray company, no reference). All the initial powders were sieved and no powders over 200 μm were used. The Mg–5 wt% X mixtures (where X represents graphite and/or Ni) were synthesized via ball milling using a Fritsch P5 ball miller. The process was carried out in a stainless steel vial under 10 bar of Ar for durations of 1, 3, and 5 hours. Each milling session consisted of 15 minutes of continuous milling followed by a 2-minute interval to allow the system to cool down. The ball-to-powder weight ratio was set at 17:1, equating to 8 grams of powder and 34 stainless steel balls (each with a diameter of 10 mm), while the rotation speed remained constant at 250 rpm. The milling vial was handled in a glove box filled with purified argon. The hydrolysis reaction was carried out in NaCl 3.5 wt% solution. The samples were characterized by X-ray diffraction (XRD) using a PANalytical X'Pert Pro diffractometer with Cu $K_{\alpha 1}$ radiation. Scanning electron microscopy (SEM) observations were made using a TESCAN VEGA3 SB microscope. Particles size were measured using Laser Granulometry (Mastersizer 2000S).

2.2 Life cycle assessment methodology

The LCA study was carried out according to recommendations of the ISO 14040 [28] and 14044 [29] standards referring to the four stages of the assessment: goal and scope definition, life cycle

inventory analysis, life cycle impact assessment, and interpretation, which are adapted and integrated in the conventional format of an academic paper structure. Goal, scope, and life cycle inventory are detailed in the current section. The life cycle impact assessment and interpretation stages are part of the results and discussion section of this article.

2.3 Goal and Scope

The goal of this study was to identify the environmental impacts and respective contributions of different reagents and processes involved in the hydrogen generation from magnesium alloys based on current manufacturing knowledge at the laboratory scale using magnesium waste in order to establish the most favorable production pathways. The system boundaries (Figure 1) of the LCA included raw material extraction, material processing, and hydrogen production using waste streams. The use- phase and end-of-life scenarios were excluded since they fall outside of the focus on the environmental performance of the production pathway and given that hydrogen is used in multiple applications. Relevant material, energy and transportation related flows and environmental emissions were considered whenever possible, whereas the impacts of capital goods such as building facilities and machinery were excluded as those could not be envisioned at the current stage of technological maturity and are generally small. The decision to exclude capital goods is supported by work conducted by Frischknecht *et al.* [30].

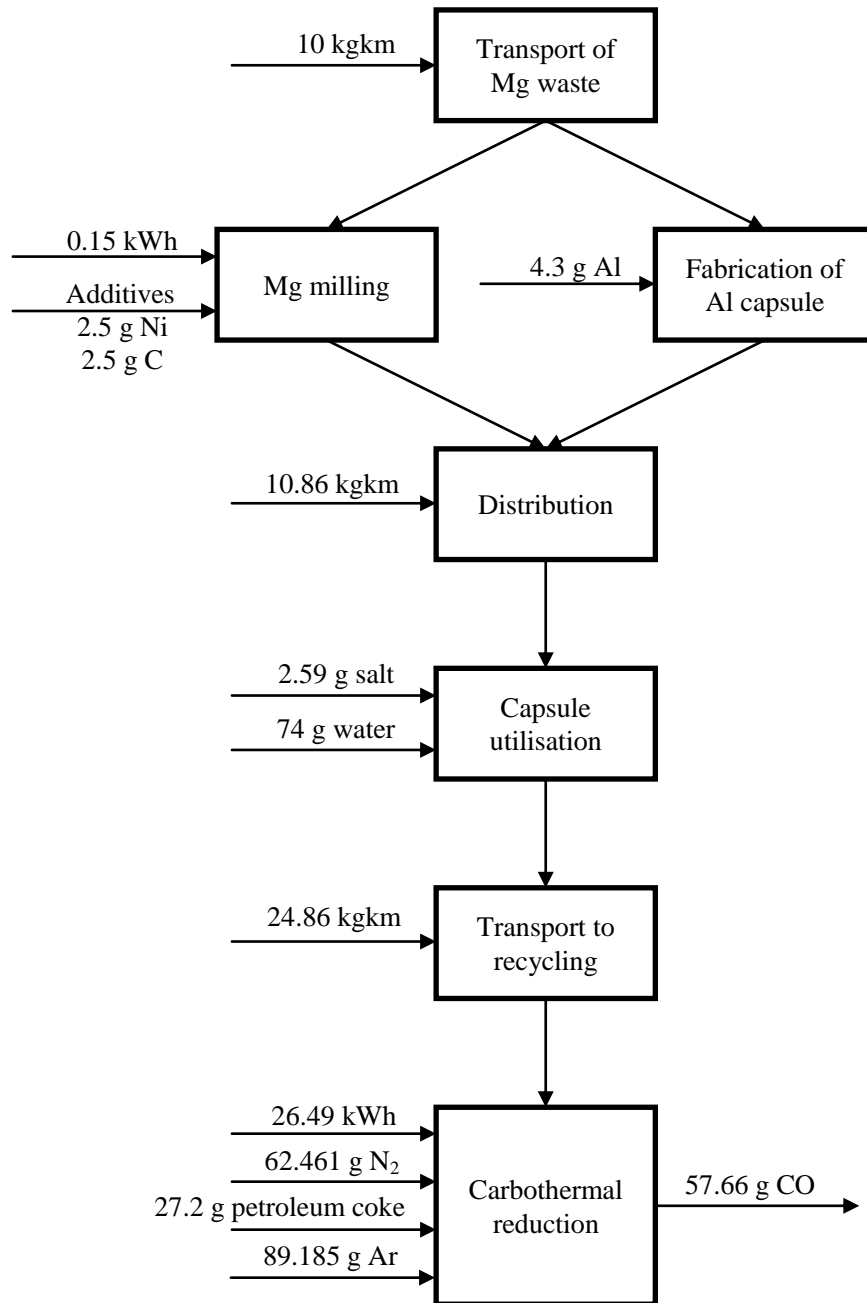


Figure 1: System boundaries and life cycle inventory flows (designed for the functional unit : a capsule of 50g of magnesium waste necessary to run a bike for 50 km)

The study was built as an ex-ante (or prospective) LCA wherein emerging technologies are analyzed at a more developed phased *e.g.* at industrial scale [31]. Prospective LCA studies are particularly useful in eco-design as they enable the optimization of emerging technologies to reduce environmental impacts [32]. Thus, the life cycle inventory analysis of the hydrogen production was established at laboratory scale but analyzed on the basis of their anticipated

occurrence in industrial settings. Compilation of life cycle inventory inputs were carried out using direct measurements at the Institute of Condensed Matter Chemistry of Bordeaux (ICMCB) of the Centre National de la Recherche Scientifique (CNRS) in Bordeaux; that means stoichiometry, thermodynamics, and expert opinion, as well as observations were derived from the processes described in the experimental part. Moreover, the hydrolysis results for H₂ production are compared with the hydrogen generation from steam reforming.

The functional unit was defined as the production of hydrogen during the total cycle of a capsule of 50g of magnesium waste (necessary to run a bike for 50 km).

The life cycle inventory model was built in the LCA software SimaPro 9, through which the life cycle impact assessment was executed. Classification and characterization of the life cycle inventory results was carried out using the ILCD endpoint method [33], including Global Warming. Principle inventory data for the hydrogen generation process were collected in-house, and foreground data were obtained from stoichiometric calculations. Modeling of background materials and processes was carried out using Ecoinvent v3.3. Electricity supply was modeled for the geographical context of France.

We present the impact assessment results primarily at the endpoint level rather than as midpoints to offer a comprehensive global perspective. Each endpoint result encompasses several midpoints, including the following:

- Human Health: human toxicity non-cancer effects, human toxicity cancer effects, particulate matter, ionizing radiation
- Ecosystem: photochemical ozone formation, acidification, terrestrial eutrophication, freshwater eutrophication, marine eutrophication, freshwater ecotoxicology
- Resources: land use, water resources depletion, mineral and fossil resources depletion

Furthermore, we specifically present the Global Warming potential, as this indicator is particularly relevant for assessing climate change.

2.4 Life Cycle Inventory

The inventory for the production of hydrogen generation from magnesium alloys using waste are given in Figure 1. The quantities of flows and emissions were determined as described in the experimental part based on stoichiometric calculations and data collections at ICMCB. The

energy required for the shredding and recycling processes was calculated directly from the equipment used. The transport distances were estimated by expert opinions based on current procurement practices for a plant in Bordeaux. Tableau 1 provides an overview of the life cycle inventory data used from Ecoinvent 3.3 for the different flows used in Figure 1.

Tableau 1: Life cycle inventory data used from Ecoinvent 3.3

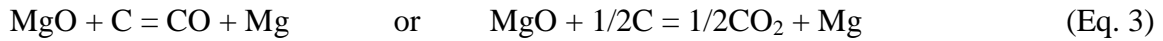
Capsule fabrication:
Aluminium, primary, cast alloy slab from continuous casting {GLO} market for Alloc Rec, U
Nickel, 99.5% {GLO} market for Alloc Rec U
Carbon black {GLO} production Allec Rec U
Electricity, medium voltage {FR} market for Alloc Rec, U
Capsule utilisation:
Tap water {Europe without Switzerland} market for Alloc Rec, U
Sodium chloride, powder {GLO} market for Alloc Rec, U
Transport:
Transport, freight, lorry 16-32 metric ton, EURO 5 {GLO} market for Alloc Rec, U
Magnesium recycling:
Electricity, medium voltage {FR} market for Alloc Rec, U
Nitrogen
Argon

A life cycle assessment of the reforming of methane by steam has already been carried out as part of a thesis [34]. The data from the thesis as well as that presented in the article by Koroneos *et al.* [35], enabled the (rough) modeling of the impact of methane steam reforming to be compared to that of the hydrolysis of magnesium, with a focus on the production routes.

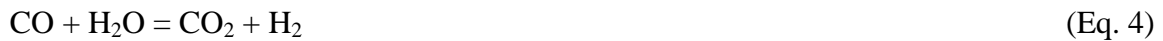
3 Results and discussion

Ensuring the green production of hydrogen poses a critical challenge for transitioning to a low-carbon H₂ economy. Over the past decade, numerous studies [12,20,23,36] have explored the hydrolysis reaction extensively. Utilizing magnesium or its hydrides to generate hydrogen through simple reactions (Equations 1 and 2) presents an affordable and straightforward method. Additionally, the use of non-recycled magnesium alloys, which constitute over half of all used Mg alloys, provides a second life for these materials [8]. However, while the resulting product of

the reaction (*i.e.*, Mg(OH)₂) is non-hazardous, its recycling remains a consideration. The commonly employed method is carbothermal reduction (CTR) [37,38]. Initially, the hydroxide undergoes nitrogen exposure at 650°C to yield the oxide (MgO) through pyrolysis. Subsequently, the CTR process occurs, with the reaction expressed as follows:



This reaction occurs at a minimum of 1500°C and under reduced pressure of argon atmosphere (typically 10 kPa). It is important to note that, given the initial reaction, carbon monoxide (CO) can also be valorized in the final reaction involved in steam reforming:



The life cycle assessment (LCA) encompasses the entire process, depicted in Figure 1, of hydrogen production from hydrolysis using recycled Mg alloys, followed by the pyrolysis of Mg(OH)₂ and the carbothermal reduction (CTR) of MgO. This assessment considers the complete magnesium cycle, focusing on the prototype described in [21], which involves a capsule made of aluminum containing the Mg alloy with Ni and C additives. The results are summarized in Figures 2 and 3.

Figure 2 clearly indicates that the environmental impacts of using the capsule, managing both waste and magnesium alloys transport, as well as delivery, are negligible according to the LCA. Conversely, the most significant impacts stem from the recycling of Mg(OH)₂. This outcome was expected because, while other processes occur at room temperature, pyrolysis and CTR involve high-temperature processes. Therefore, recycling of Mg(OH)₂ should be considered through other way than CTR to have a lower impact. Some research are conducted using solar furnace but chemical routes could also been considered. Figure 3 provides detailed insights into the impacts of this recycling, highlighting that thermal treatment and pyrolysis account for approximately 90% of the total impact across various metrics. Therefore, to ensure that H₂ production through hydrolysis is environmentally friendly (*i.e.*, no CO₂ emission, no use of acid or basic solution, no addition of critical raw materials), it is imperative to adopt a green method for recycling Mg(OH)₂. Some studies propose using solar furnaces to reduce environmental impacts [26].

The impacts of the capsule process itself average around 10% of the total impact. A closer examination reveals that impacts on ecosystems and global warming are relatively limited (up to 5% maximum), with the highest impact observed on human health (12%).

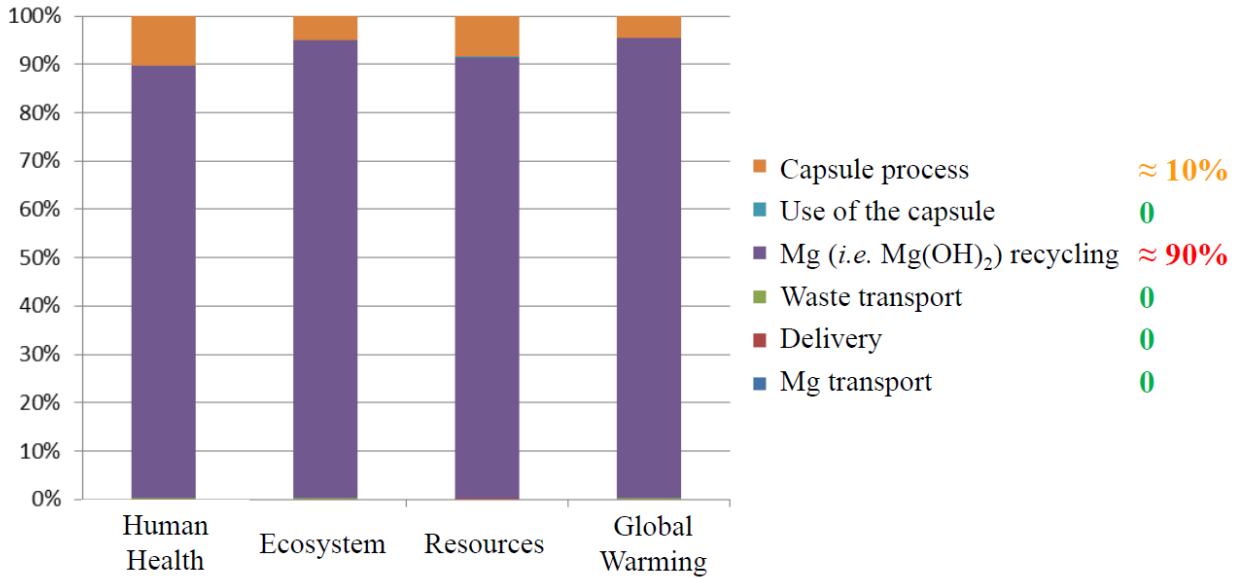


Figure 2: The environmental impacts of the total cycle of a capsule

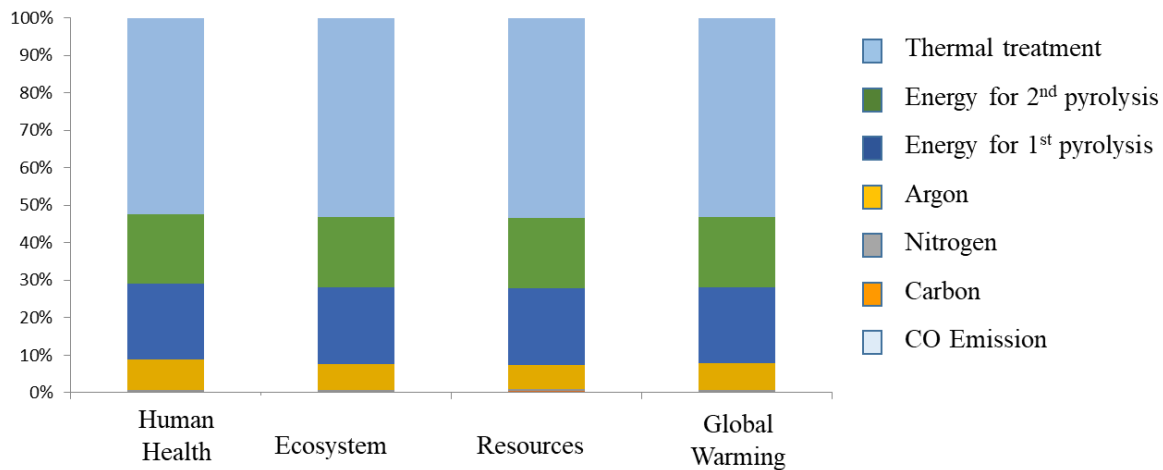


Figure 3: The environmental impacts of the recycling of Mg(OH)₂

To gain more insight into the specific environmental impacts of H₂ production, we conducted a separate life cycle assessment (LCA) focusing solely on the Mg capsule. The findings are outlined in Figure 4a (left figure). Ni and Al together were the primary contributors across all metrics; Ni had the most significant impact on Human Health and Resources, while Al had the greatest impact on Ecosystem and Global Warming. C contributed less than 10% across all metrics. One potential solution to mitigate these impacts is to use recycled Al instead of virgin

Al. Consequently, we conducted a revised LCA considering recycled Al, and the results are depicted in Figure 4b (right figure). Considering this new scenario, the relative impact of Al is notably reduced, while Ni remains a major contributor. Nevertheless, it appears that (i) the relative contribution of Al capsule fabrication increases in terms of impacts on Ecosystem and Global Warming and (ii) the relative contributions of milling and C increase across all metrics. Both scenarios' results highlight the necessity to address the impacts of Ni to enhance overall performance.

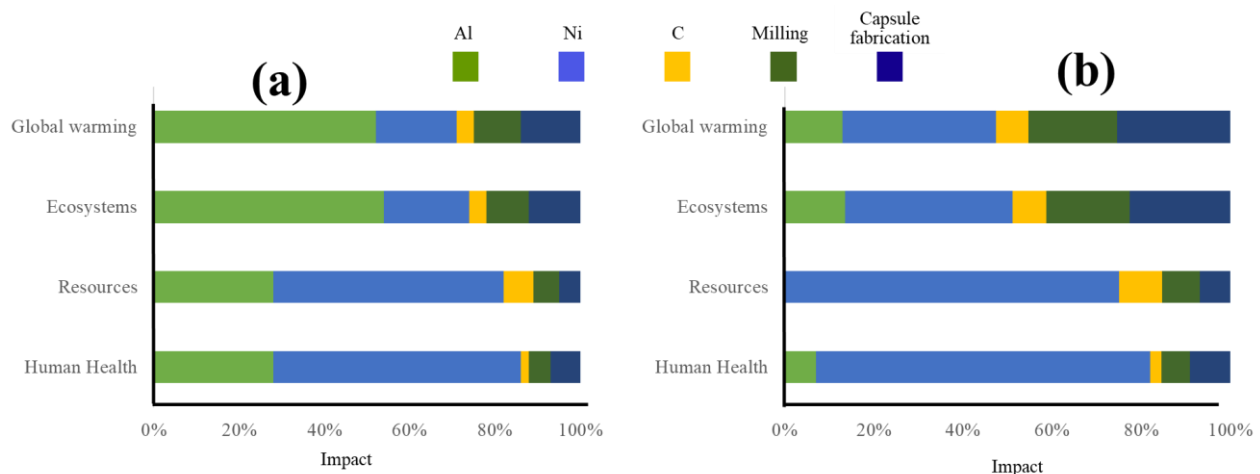


Figure 4: The impacts of the cycle of the « Mg » capsule only. (a) considering pure Al and (b) considering recycle Al.

The outcomes from various Mg combinations are detailed in Figure 5. It is evident that using pure magnesium or incorporating Al does not yield more than 20% even after 15 minutes. However, the addition of transition metals such as Ni and Fe results in a significantly higher yield within about 5 minutes (80% and 60%, respectively). This effect is primarily attributed to the galvanic coupling between magnesium and the transition metal, with a stronger coupling observed in the case of Ni due to a higher Open Circuit Potential (OCP) difference [16].

The significance of carbon (C) is underscored, explaining its role in terms of (i) acting as a lubricant during milling, thus enhancing milling process efficiency, (ii) forming a protective layer on the magnesium surface, and (iii) establishing a galvanic coupling between magnesium and carbon [23]. These factors collectively contribute to a 100% yield. Similarly, a 100% yield is

achieved by mixing magnesium with both Ni and C (Mg + 5wt.%C + 5wt.%Ni). In this scenario, a 100% yield is attained in less than 3 minutes.

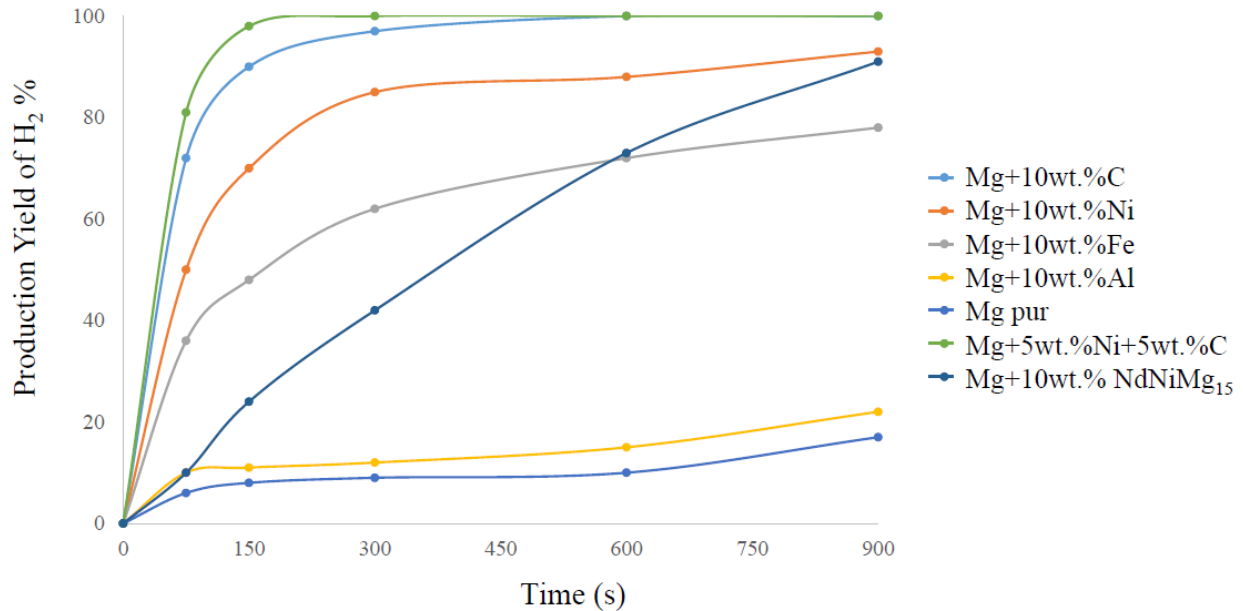


Figure 5: Hydrogen generation yield obtained by hydrolysis with several magnesium mixtures (e.g., a yield of 100% correspond roughly to 1L of H₂/g of Mg as all the mixtures contain the same amount of Mg, expected the Mg pur in dark blue)

Despite achieving a high yield, the results of the LCA study indicate a need to avoid or minimize the use of nickel (whatever the form of nickel). Consequently, exploring the addition of binary and ternary compounds containing Ni becomes feasible. Through this approach, El Asnar et al. achieved outstanding results by introducing NdNiMg₁₅ [36]. The reported results are based on small bulk samples, which may account for the relatively lower kinetic value observed, although the yield was nearly 100% within just 15 minutes.

Furthermore, utilizing bulk samples not only eliminates the need for the milling process but also addresses the relatively limited impact observed in the LCA results. Consequently, even though the milling's impact was limited, it will be suppressed in this context. Additionally, handling bulk materials is less hazardous and more manageable compared to handling powders.

Using NdNiMg15 as an additive instead of pure Ni, with equal weight percentages of 10% in each case, allows for the reduction of the total weight percentage of nickel in the material to approximately 1%. Consequently, the environmental impact of nickel in the life cycle assessment (LCA) will be reduced by a factor of 10 compared to the impact shown in Figure 4. However, it is crucial to also consider the environmental impacts of rare earth elements and the synthesis of ternary compounds in this scenario.

This reduction in nickel content signifies viable solutions for drastically decreasing the amount of Ni used. However, it also implies the necessity of evaluating the impacts of rare earth elements and ternary compound synthesis.

The environmental impacts related to steam reforming, as modeled in [34] and [35], are likely underestimated due to the exclusion of certain parameters. Conversely, the data regarding hydrolysis represent a comprehensive assessment, considering all relevant parameters. Therefore, any comparison should be conducted cautiously, and it may be necessary to conduct a thorough reevaluation of the steam reforming process to gain accurate insights.

The findings of this comparison are illustrated in Figure 6a and Figure 6b. According to Figure 6a, the environmental impacts of the hydrolysis reaction for hydrogen production are roughly half those of steam reforming in terms of Ecosystem and Global Warming. This observation aligns with the assertions of numerous authors [8, 12-14, 16-23] who advocate for hydrolysis as an environmentally friendly method for hydrogen production. However, it is essential to note that the impacts on Human Health and Resources are respectively 3 and 10 times greater for hydrolysis compared to steam reforming. Despite potential factors not being fully considered for steam reforming, these results strongly suggest the need for a reassessment of hydrolysis as currently practiced.

As depicted in Figure 6b, the primary concern lies in the impact of nickel on Human Health, accounting for roughly 80%. Mitigating the impact of nickel would significantly reduce the Human Health impact (~25% less) for hydrolysis. Surprisingly, even with nickel removed from the equation, the impact on Resources remains higher for hydrolysis than for steam reforming. However, this discrepancy stems from the methodology of accounting for Resource impacts in

the ILCD endpoint method, which heavily relies on the depletion approach. This approach has faced scrutiny and differing interpretations among scholars [39–43].

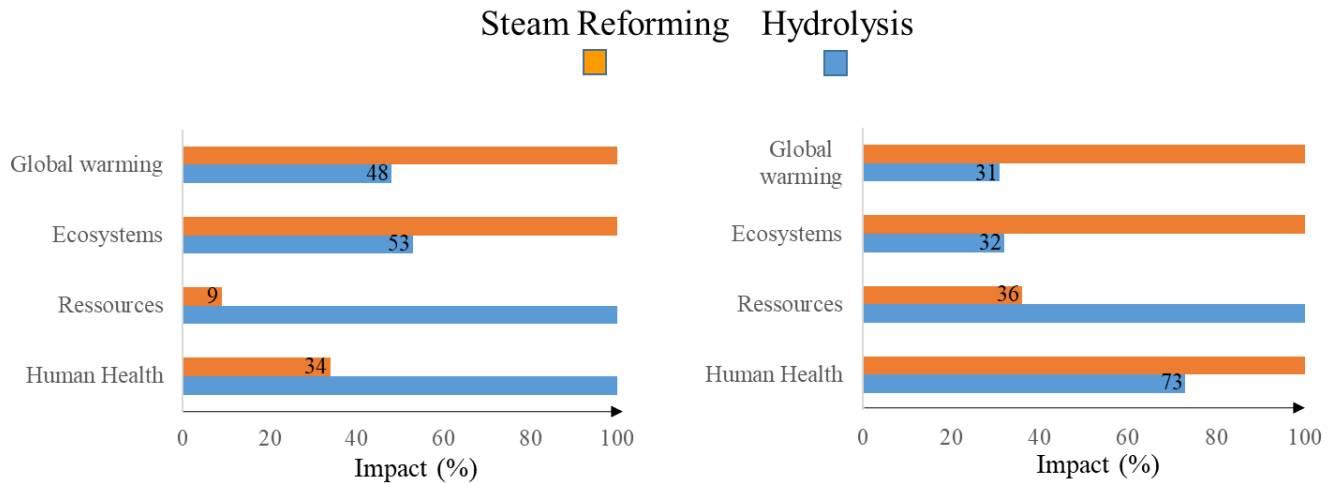


Figure 6: The environmental impacts of hydrogen production by steam reforming and by hydrolysis [34,35]. On the left (Figure 6a), the hydrolysis process takes into account the nickel and on the right (Figure 6b), the impact of nickel has been taken off of the LCA study

4 Limitations

It is important to highlight the limitations of this study. The study was conducted at a laboratory level therefore, it can only be considered an indication of performance at an industrial scale. Once the processes have been moved to a higher TRL, it would be worth redoing the LCA study.

5 Conclusions

Firstly, we have demonstrated through an LCA study that considering the recycling of $Mg(OH)_2$ using the classical carbothermal reduction (CRT) method is not environmentally sound, as it contributes to nearly 90% of the total environmental impacts. Secondly, in exploring hydrogen production through hydrolysis with magnesium, it becomes evident that aluminum and nickel significantly influence environmental impacts. Thus, efforts should be made to mitigate their effects. Utilizing any type of recycled aluminum instead of pure aluminum offers a straightforward solution to reduce aluminum's environmental impacts. To address nickel, exploring ternary magnesium-based compounds could be a viable option.

Thirdly, our findings indicate that hydrolysis with magnesium is advantageous in terms of Ecosystem and Global Warming impacts and is favorable for Human Health (when nickel is avoided). However, it remains 3 to 10 times worse than steam reforming concerning Resources impacts. It is crucial to note that the assessment of resource impacts in the applied impact assessment method heavily relies on the depletion approach, which has significant variability depending on interpretation.

Overall, these results highlight the potential environmental superiority of the hydrolysis process over steam reforming. However, they also underscore the need for further environmentally friendly design of these processes, especially in the context of promoting a circular economy. Consequently, it can be affirmed that, at present, employing hydrolysis with magnesium represents a relatively eco-friendly approach compared to steam reforming. However, scaling up from laboratory-scale operations is imperative; the current state of the technology is not yet suitable for large-scale hydrogen production and should primarily be considered for niche applications.

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