LCA FOR ENERGY SYSTEMS AND FOOD PRODUCTS

Life cycle assessment of hydrogen energy systems: a review of methodological choices

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Abstract

Purpose As a first step towards a consistent framework for both individual and comparative life cycle assessment (LCA) of hydrogen energy systems, this work performs a thorough literature review on the methodological choices made in LCA studies of these energy systems. Choices affecting the LCA stages "goal and scope definition", "life cycle inventory analysis" (LCI) and "life cycle impact assessment" (LCIA) are targeted.

Methods This review considers 97 scientific papers published until December 2015, in which 509 original case studies of hydrogen energy systems are found. Based on the hydrogen production process, these case studies are classified into three technological categories: thermochemical, electrochemical and biological. A subdivision based on the scope of the studies is also applied, thus distinguishing case studies addressing hydrogen production only, hydrogen production and use in mobility and hydrogen production and use for power generation.

Results and discussion Most of the hydrogen energy systems apply cradle/gate-to-gate boundaries, while cradle/gate-tograve boundaries are found mainly for hydrogen use in

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mobility. The functional unit is usually mass- or energybased for cradle/gate-to-gate studies and travelled distance for cradle/gate-to-grave studies. Multifunctionality is addressed mainly through system expansion and, to a lesser extent, physical allocation. Regarding LCI, scientific literature and life cycle databases are the main data sources for both background and foreground processes. Regarding LCIA, the most common impact categories evaluated are global warming and energy consumption through the IPCC and VDI methods, respectively. The remaining indicators are often evaluated using the CML family methods. The level of agreement of these trends with the available FC-HyGuide guidelines for LCA of hydrogen energy systems depends on the specific methodological aspect considered.

Conclusions This review on LCA of hydrogen energy systems succeeded in finding relevant trends in methodological choices, especially regarding the frequent use of system expansion and secondary data under production-oriented attributional approaches. These trends are expected to facilitate methodological decision making in future LCA studies of hydrogen energy systems. Furthermore, this review may provide a basis for the definition of a methodological framework to harmonise the LCA results of hydrogen available so far in the literature.

Keywords Energy system . FC-HyGuide . Hydrogen . Life cycle assessment . Methodology . Trend

1 Introduction

The global energy demand continues to grow in order to satisfy higher standards of living and allow the world economy to thrive. Fossil fuels are the dominant source in the current energy context, and they are expected to supply 80 % of the total primary energy demand by 2035. Nevertheless, the use of renewable sources is set to grow rapidly, and their demand is forecasted to increase fourfold by 2035 (British Petroleum [2016\)](#page-14-0). In particular, hydrogen could play a significant role in achieving sustainability targets according to future energy scenarios (International Energy Agency [2014](#page-15-0)).

Hydrogen arises as a carbon-free energy storage option with high energy content per unit mass (Suleman et al. [2015\)](#page-16-0) and different storage techniques (mainly, compressed, liquefied or adsorbed in metal hydrides). These features make hydrogen interesting for decarbonising the energy system (Marchetti [2005](#page-15-0); Dunn [2002](#page-14-0); International Energy Agency [2014;](#page-15-0) Sgobbi et al. [2015](#page-16-0)). Transport is seen as the key field of application for hydrogen because of the need for lowcarbon fuels to gradually replace conventional fuels and thereby minimise life cycle emissions over the supply chain. Hydrogen produced from fossil resources with $CO₂$ capture, hydrogen by electrolysis with low-carbon power and hydrogen by biogas reforming or biomass gasification are among the options considered for this shift in transportation fuels (International Energy Agency [2014](#page-15-0), [2015](#page-15-0)). However, hydrogen is currently associated mainly with non-energy applications such as ammonia production (Simons and Bauer [2011](#page-16-0); Bhandari et al. [2014](#page-14-0)). In fact, important challenges have to be faced to make the development of a hydrogen economy feasible, including not only techno-environmental issues but also concerns about social acceptance (Iribarren et al. [2016](#page-15-0)) and cost-competitive roll-out of technological solutions and infrastructure for distribution and use (Dincer and Acar [2015](#page-14-0)). Regarding techno-environmental issues, since hydrogen is not directly available in its pure form but bound to other elements, high energy expenditure is needed to separate hydrogen. Hence, the environmental and energy performances of hydrogen energy systems strongly depend on the hydrogen donor and the energy source for the conversion process (Serrano et al. [2012;](#page-16-0) Martín-Gamboa et al. [2016\)](#page-15-0). In this sense, comprehensive analyses are required in order to evaluate the environmental suitability of hydrogen energy systems.

Because life cycle assessment (LCA) is a well-established methodology for the comprehensive evaluation of the potential environmental impacts of product systems, a relatively high number of LCA studies of hydrogen energy systems can be found in the scientific literature. Despite international standardisation of LCA (ISO [2006a](#page-15-0), [b](#page-15-0)) and the availability of both general and specific LCA guidance documents (JRC [2010;](#page-15-0) Lozanovski et al. [2011;](#page-15-0) Masoni and Zamagni [2011](#page-15-0)), each of the LCA studies of hydrogen energy systems follows its own methodological choices. This strongly affects the results of the LCA studies and their final interpretation. Furthermore, the differences in methodological choices and other technical parameters prevent a robust comparison of the results from different studies (Bhandari et al. [2014](#page-14-0)). Within this context, this article presents a thorough review

of the methodological choices made in LCA studies of hydrogen energy systems. The identification and analysis of trends and singularities in methodological choices is expected to be of paramount importance not only for the performance of future LCA studies of hydrogen energy systems but also for the future definition of a methodological framework to harmonise LCA results of this type of product system. This is deemed possible thanks to the high level of detail attained in the present review. Hence, this study aims to reveal methodological differences in LCA of hydrogen energy systems and pave the way for a more robust and standardised protocol for both individual and comparative LCA studies. Finally, it should be noted that this study is conducted within the frame of Task 36 of the International Energy Agency (IEA) Hydrogen Implementing Agreement (HIA). The goal of IEA HIA Task 36 is to facilitate decision-making in the hydrogen energy sector by providing a robust methodological framework based on the life cycle sustainability assessment (LCSA) of hydrogen energy systems. In this respect, LCSA (Guinée et al. [2011](#page-15-0); Sala et al. [2013\)](#page-16-0) arises as a comprehensive framework for assessing economic, social and environmental aspects of hydrogen energy systems.

2 Materials and methods

This work presents a review of LCA studies of hydrogen energy systems available until December 2015. Only published articles (Scopus search; keywords "hydrogen" AND "life cycle assessment") and key reference reports released by well-known research laboratories are taken into account. All these works include at least the hydrogen production stage within the system's boundaries. Studies with a cradle-to-grave scope as well as studies with a more limited scope are therefore included. For the sample of LCA studies, each variation in technological aspects (e.g. feedstock, energy carrier, and hydrogen storage technology) or methodological decisions (e.g. functional unit, system boundaries, reference region, and allocation approach) defines a new (original) case study for the purpose of this review. Case studies dealing with projected scenarios for sensitivity analysis are not considered to be new case studies. In order to avoid potential distortion of the observed trends, articles with a very high number of different case studies (>30) are not included in the literature analysis. Moreover, those case studies considering hydrogen as a by-product from a background process are excluded inasmuch as the hydrogen production phase is set as a mandatory component of the study. When dealing with groups of articles based on the same hydrogen energy systems with the same methodological choices and published by the same authors, only the most representative article is taken into consideration. Methodological choices in three of the four LCA stages are addressed in this review. These stages are goal

and scope definition, life cycle inventory analysis (LCI), and life cycle impact assessment (LCIA). The case studies reviewed are subdivided in terms of system boundaries, functional unit (FU), allocation approach, data sources, impact categories, and impact assessment method. The trends observed for these issues are contextualised by comparison with the corresponding recommendations provided by the FC-HyGuide guidance document for performing LCA on hydrogen production systems (Lozanovski et al. [2011\)](#page-15-0).

After the screening of published works, the final sample includes 97 publications (Tables [1](#page-3-0) and [2](#page-4-0)), in which 509 original case studies are identified. Based on the classification proposed by Dincer [\(2012](#page-14-0)) and Dincer and Zamfirescu [\(2012\)](#page-14-0), the case studies are divided into three technological categories: thermochemical, electrochemical and biological hydrogen production. Regarding the thermochemical category, the hydrogen production technologies found are: biomass or coal gasification, steam reforming of fossil fuels or biofuels, partial oxidation of fossil hydrocarbons or biofuels, thermal cracking of fossil hydrocarbons and water splitting based on thermochemical cycles. The technologies found for the electrochemical category are low- and high-temperature electrolysis. Additionally, water photosplitting and plasma arc decomposition of natural gas are approached as technologies belonging to the electrochemical category. Finally, the technologies found for the biological category are: dark, photo and two-step fermentation of biomass or microalgae and biophotolysis led by microalgae.

Tables [1](#page-3-0) and [2](#page-4-0) gather the LCA studies of hydrogen energy systems included in this review and published before and after the release of the FC-HyGuide guidance document, respectively. The column "region and base year" refers to the specific geographical coverage and year or period considered in the study, when reported. In the column "scopes", the label "XtoGate" is used for the case studies that do not include the hydrogen use stage (i.e. the "grave") within the system's boundaries. In these case studies, "X" means either the feedstock production stage or the production of the energy source driving the conversion stage, while "gate" means hydrogen production, purification, conditioning or distribution. Regarding the "number of original H_2 case studies", it should be noted that, in comparative LCA studies, only the case studies whose results are not directly taken from previous studies are considered to be original.

Figure [1](#page-5-0) provides further bibliometric details about the sample of studies reviewed. In this figure, unlike in Tables [1](#page-3-0) and [2,](#page-4-0) "region" refers to the region of the first author's affiliation. The activity trend shown in Fig. [1](#page-5-0) is generally consistent with that provided by Bhandari et al. (2014) (2014) in a similar study. An increasing interest in LCA of hydrogen energy systems (hydrogen production and/or use) is observed, with more than 60 % of the publications embraced in the period 2011–2015. Regarding the focus of the study, works focused on hydrogen production are distinguished from those focused on hydrogen use in mobility or stationary applications (stationary means hydrogen-to-power and, to a lesser extent, hydrogen-to-fuel studies). In this respect, 51 % of the publications deal with hydrogen production, 39 % with hydrogen use (33 % mobility and 6 % stationary), and the remaining 10 % with both hydrogen production and use. The analysis of the affiliation region can give an insight into regional differences in the focus of the study. In this sense, the studies with European or North American affiliation focus mainly on hydrogen production, whereas the studies coming from Asia are focused mainly on hydrogen use for mobility application.

3 Results and discussion

3.1 Goal and scope definition

All the studies included in the review involve an attributional approach. Hwang [\(2013\)](#page-15-0) presents the only study of hydrogen from first-generation biomass evaluating explicitly greenhouse gas (GHG) emissions from land use change (using the GREET model (Wang [1996](#page-16-0))), but the study is not stated as a consequential LCA. In fact, consequential LCAs of hydrogen energy systems are not yet available in the literature in a number allowing a specific review. An example of consequential LCA of hydrogen technology is presented by Chen et al. [\(2012a](#page-14-0), [2012b](#page-14-0)), but their work is excluded from this review since it is focused on a "hydrogen society" based on different hydrogen technologies mutually interacting rather than on a specific hydrogen-production supply chain.

The first stage of the LCA framework defines key aspects such as the system's boundaries and the FU, as well as the objectives and potential uses of the study. Around 90 % of the studies reviewed are comparative, with 80 % of them presenting robust (i.e. consistent with the methodological framework) comparisons, mainly because the systems compared come from the same study. On the other hand, in the remaining comparative studies, the comparison with the life cycle performance of systems assessed in other studies is not generally performed using consistent system boundaries.

3.1.1 Functional unit

The FU quantifies the function of the product system and provides a reference unit. The choice of the FU can strongly affect the conclusions of the study (especially in comparative studies) and must be defined in accordance with the goal and scope of the study. Figure [2](#page-6-0) shows the nature (mass, volume, energy, travelled distance, passenger·distance or economic) of the FU chosen according to the type of case study under review. Overall, the choice of FU is found to be similarly distributed between energy, mass and travelled distance. In

Reference Scope Region and base year No. original H₂

Table 2 List of life cycle studies reviewed: years 2012–2015

Table 2 (continued)

^a Study available online in 2015 despite final publication in 2016

hydrogen production case studies (XtoGate), the common choice is an FU that allows a direct identification of an amount of hydrogen. It should be noted that a small percentage of case studies which use an energy FU do not present a clear statement about the energy content (lower or higher heating value), while case studies using a volume FU always specify appropriately pressure and temperature conditions (necessary to convert volume into mass or energy). Economic FUs play a minor role, accounting for only four case studies presented in the same article (Pacheco et al. [2015](#page-16-0)). Moreover, in that work, the authors do not use exclusively an economic FU, but they present a comparison showing how the choice of economic,

Fig. 1 Overview of the sample of publications reviewed

mass or energy FU, combined with different allocation approaches, influences the final interpretation of the results in a biological multifunctional system. Input-oriented FUs are used in a minor number of case studies related to a multifunctional system using an FU based on the energy content of the feedstock (Tock and Maréchal [2012](#page-16-0)).

According to the FC-HyGuide guidance document, the FU of hydrogen production systems should be defined as "a certain amount of hydrogen produced^ and complemented with information on energy content or thermodynamic conditions. In this respect, the FU trends observed for this type of system are in line with FC-HyGuide recommendations.

Regarding the case studies involving hydrogen use in stationary applications, all authors use an energy FU consistent with the function of the target systems (energy production). Regarding the case studies addressing mobility applications, the most common FU choice is "travelled distance", being "passenger·distance" a less used FU. Ramos Pereira et al. (2014) show the effect of changing the FU ("km travelled" and "passenger-km") in an XtoGrave study for the aviation sector. Three case studies use the exergy content of hydrogen as FU (Granovskii et al. [2007](#page-15-0)); these case studies are embedded in the energy-FU category in Fig. 2. Finally, the case studies that use an energy FU for mobility refer to the energy content of hydrogen.

3.1.2 System boundaries

In this review, the inclusion of the hydrogen production stage—in which the hydrogen donor (green or fossil) is converted into high-purity hydrogen—is set as a mandatory component within the system's boundaries of each case study. The energy source drives this conversion stage, which is based on electrochemical, thermochemical, biochemical, radiochemical or photochemical technology (Dincer [2012](#page-14-0); Dincer and Zamfirescu [2012](#page-14-0)). Only processes belonging to the thermochemical, electrochemical and biological categories are found in this review.

Figure [3](#page-7-0) shows the number of case studies broken down by specific stages within the system's boundaries, also showing technological choices and inclusion of capital goods (i.e. goods such as machinery, equipment and buildings) at each stage. Based on FC-HyGuide, the stages considered in this figure are: hydrogen production, purification, storage, distribution and use. The end-of-life stage of hydrogen energy systems is currently underdeveloped, which is being currently faced within the framework of an international project funded by the Fuel Cells and Hydrogen Joint Undertaking (HYTECHCYCLING project, Grant Agreement No. 700190). The number of XtoGrave case studies corresponds to the number of cases that include hydrogen use (226), while the resultant number of XtoGate case studies is 283 (the difference between the total number of case studies [509] and the number of XtoGrave case studies [226]). Among the studies focused on hydrogen production, the production stage is considered the final "gate" in 16 % of them, mainly in cases of hydrogen produced through thermochemical cycles and electrolysis. Nevertheless, the purification step is found to be the most common final gate (36 % of the XtoGate case studies), mainly in cases of hydrogen production through thermochemical and biological processes. Electrochemical XtoGate cases, in contrast, mainly consider the conditioning stage (compression, liquefaction or solid storage in metal hydrides) as the final gate (79 % of those cases). An additional stage that can be considered in XtoGate case studies is hydrogen distribution. A significant number of XtoGate case studies assume this option as the final gate (29 %). However, this is linked to "well-to-wheels" (i.e. cradle-to-grave) studies in which the

Fig. 3 Technical choices according to the life cycle stage (white dots represent case studies that include capital goods)

overall boundaries are subdivided into "well-to-tank" (i.e. cradle-to-gate; considered XtoGate herein) and "tank-towheels" (i.e. gate-to-grave).

Regarding the hydrogen production stage (found in all cases, i.e. 509 case studies), the most assessed process in the thermochemical category is steam reforming (SR; 54 % of the thermochemical case studies, and 30 % of the total number of case studies). This is closely linked to the fact that most of the studies are comparative and usually take conventional SR of natural gas (steam methane reforming, SMR) as the reference technology. Apart from SR, the most studied thermochemical technologies are gasification and thermochemical cycles. These processes are found in 26 and 15 % of the thermochemical case studies, respectively (15 and 9 % of the total amount of case studies). Partial oxidation, thermal cracking and autocatalytic decomposition complete the variety of thermochemical technologies found. It should also be noted that, in the thermochemical category, 10 % of the case studies consider $CO₂$ capture (37 % of them in coal gasification, 30 % in biomass gasification, 26 % in SMR and 7 % in natural gas splitting by chemical looping). The electrochemical category is clearly dominated by water electrolysis (mainly alkaline and, in a lower number of cases, high-temperature, highpressure and proton exchange membrane electrolysis). Only three and two case studies deal with plasma arc decomposition of methane (Wagner et al. [1998](#page-16-0); Pehnt [2003\)](#page-16-0) and water photosplitting (Dufour et al. [2012\)](#page-14-0) respectively. In the biological category, 90 % of the case studies are based on fermentation (photo and dark sequentially, photo only or dark only) and 10 % on bio-photolysis (carried out by microalgae).

Regarding the purification stage (found in 289 case studies), most of the case studies consider pressure swing adsorption (PSA), which is the conventional process used in the thermochemical category (SR and gasification, mainly) to meet the H_2 -purity requirement. In electrochemical technology, purity higher than 99.9 % can be achieved without a purification step. Pd membrane and deoxygenation processes are other purification technologies found in the review. The former is considered by Chang et al. ([2007](#page-14-0)) after SR while the latter is added to the electrolysis step in order to reach H_2 purity above 99.999 % (Lombardi et al. [2011\)](#page-15-0). These two purification processes are included in the category "other" in Fig. 3, together with those cases for which the separation process is not clearly specified (actually, the unspecified cases are found to dominate the category "other").

Storage (found in 329 case studies) can take place after and/ or before the distribution stage (when included). In those case studies including both storage and distribution steps, only a small percentage of the cases (13 %) apply storage both before and after distribution, with the remaining cases showing similar contributions between storage before or after distribution. The labels "adsorbed" (in metal hydrides), "liquefied" and "compressed" (gas) in Fig. 3 refer to the final state of the hydrogen. The most common choice is compressed hydrogen. The pressures assumed are within the range 80–880 bar for mobility use and 8–70 bar for hydrogen production. When compared to liquid and compressed hydrogen, capital goods (white dots in Fig. 3) are more often included for hydrogen stored in the solid matrix of metal hydrides. Several authors present studies evaluating the differences in impacts and energy consumption when considering alternative storage options (Wagner et al. [1998;](#page-16-0) Chang et al. [2007](#page-14-0); Hwang and Chang [2010;](#page-15-0) Ramos Pereira and Coelho [2013](#page-16-0); Sevencan and Çiftcioglu [2013](#page-16-0); Muresan et al. [2014](#page-15-0); Tong et al. [2015](#page-16-0)).

Regarding the distribution stage (found in 298 case studies), transportation by lorry (in liquid or compressed gas state) and pipeline distribution show significant contribution percentages in Fig. 3. Hydrogen distribution is classified as "not needed" for all the cases involving stationary applications as well as when hydrogen is produced directly in the refuelling station.

Proton exchange membrane fuel cell (PEMFC) technology is the most common option in hydrogen use, for both mobility and energy production. In mobility applications, hydrogen as a fuel in vehicles with internal combustion engine is considered in less than 10 % of the case studies. Regarding stationary application, 3 out of 27 cases use hydrogen for methanation via the Sabatier reaction (Reiter and Lindorfer [2015\)](#page-16-0), while the remaining cases (24) address power production: 2 cases using molten-carbonate fuel cell technology (Lunghi et al.

[2004\)](#page-15-0), 15 using PEMFC technology, and 7 using a gas turbine. Pehnt [\(2003](#page-16-0)) also considers solid oxide fuel cells but without using hydrogen as the fuel.

Key aspects determining the life cycle performance of hydrogen production systems are the source of energy driving the hydrogen production process and the raw material that contains hydrogen (Serrano et al. [2012](#page-16-0); Susmozas et al. [2016\)](#page-16-0). On the one hand, since the electrochemical category is widely dominated by case studies of water electrolysis, the environmental performance of this type of system strongly depends on the energy source. On the other hand, the performance of biological and thermochemical technologies (except for thermochemical cycles) depends mainly on the hydrogen donor. Figure 4 shows the choice of power source in electrochemical case studies and the choice of hydrogen donor in thermochemical and biological case studies.

Most of the electrochemical case studies (60 %) are based on renewable sources. Nevertheless, a significant percentage of non-renewable sources is observed (40 %), mainly because most of the studies carry out comparisons between renewable and non-renewable hydrogen energy systems. Electricity (grid) mixes are considered herein "non-renewable" regardless of the share of renewables in the grid mix. In Fig. 4, the label "solar" involves electrolysis via solar photovoltaics (PV) or solar thermal power and water photosplitting, as well as two case studies with a mix of wind and solar PV power as the energy source. The label "cogeneration" involves electricity from coal or natural gas. The cases of high-temperature electrolysis are based on nuclear energy, occasionally supported by renewable electricity. Regarding the cases based on wind power, a distinction between on- and off-shore is not possible since this aspect is often unspecified. The high percentage found for grid electricity is partly due to the common consideration of electrolysis driven by electricity from a given regional mix as a reference process.

Regarding the hydrogen donor for thermochemical processes (Fig. 4), natural gas is found to be the most common one. This observation matches the expectations since SMR is currently the conventional process to produce hydrogen and most of the studies are comparative and use SMR as the reference case. Moreover, natural gas is the hydrogen donor not only in SMR but also in other thermochemical processes such as partial oxidation, autothermal reforming and catalytic decomposition. Regarding "green" hydrogen donors, 10% of the thermochemical case studies are based on biofuel reforming. These biofuels come from the previous processing of different feedstocks: energy plantations (44 %), byproducts from other product systems (36 %) and municipal waste (20 %). The second most common thermochemical process is gasification, which uses as solid feedstock mainly second-generation biomass (54 %) and coal (40 %). $CO₂$ capture is included in 25 % of the case studies based on gasification.

The thermochemical case studies using exclusively water as the hydrogen donor (16 %) are based on thermochemical cycles. In this process, which takes place at temperatures significantly lower than those for water thermolysis (single-step process), water is thermally dissociated in H_2 and O_2 through different reaction steps. The reactants are regenerated and recycled at the end of the cycle, and no membrane is needed to separate H_2 and O_2 . More than 50 % of these case studies are hybrid thermochemical cycles (i.e. requiring external electricity), mainly Cu–Cl-based cycles (3-, 4- or 5-step cycle). In a significant percentage of case studies based on thermochemical cycles (13 %), the type of cycle is not stated. Concerning the energy source, the thermochemical cycles found are based on nuclear power (86 %) and, to a lesser extent, concentrated solar power (14 %).

The last pie diagram in Fig. 4 provides information about hydrogen donors considered in biological case studies. The biological technologies considered are fermentation (photo, 51 %; dark, 19 %; two-stage, 30 %) and biophotolysis. Microalgae and first- and second-generation biomass are the hydrogen donors found in the processes based on fermentation, while water is the source of hydrogen in bio-photolysis. In particular, most of the cases based on fermentation use microalgae as the feedstock, with a minor number of cases using residual organic material as the feedstock (e.g. wheat straw or potato peels).

Other general observations regarding system boundaries are: (i) the inclusion of feedstock extraction/production is a common choice in thermochemical processes based on reforming and gasification, whereas capital goods are often excluded; (ii) electrochemical and thermochemical systems based on nuclear energy often include capital goods for the nuclear plant, as well as the extraction/conversion/enrichment of uranium; (iii) power generation and capital goods are commonly included in the boundaries for electrochemical case studies based on renewable sources; (iv) a common choice for case studies addressing the use of hydrogen in mobility is the inclusion of capital goods and end-of-life for the vehicle, while capital goods linked to the fuel before use are included in a lower percentage of cases; and (v) in line with FC-HyGuide, system boundaries consistent with the goal of the study are found for all studies.

3.1.3 Multifunctionality

This section deals with choices on allocation rules for those case studies based on multifunctional systems. Approximately 19 % of the total amount of case studies are considered multifunctional. Even though thermochemical processes involve 69 % of these cases, the technological category mainly affected by multifunctionality is the biological one (50 % of the biological cases are multifunctional), followed by the thermochemical (23 %) and electrochemical (8 %) ones. The co- and by-products considered in the multifunctional case studies are mainly electricity (40 %), food and fodder (34 %), heat (22 %) and fuels (4%) . $CO₂$ capture is often included in the evaluated systems, but in all these cases, no impact is allocated to the $CO₂$ captured, and therefore, these systems are not considered multifunctional. Additionally, a significant percentage of the multifunctional case studies (37 %) present the results for the whole system (i.e. without application of any allocation approach) or do not specify clearly the allocation approach; for the remaining case studies (63%) , the choices to deal with multifunctionality are summarised in Fig. [5.](#page-10-0)

Case studies presenting multifunctionality in two different stages (Manish and Banerjee [2008;](#page-15-0) Galera and Gutiérrez Ortiz [2015;](#page-14-0) Giraldi et al. [2015](#page-15-0)) are double counted in Fig. [5](#page-10-0). System expansion is found in half of the cases with multifunctionality at the hydrogen production stage. In these systems, electricity or heat co-production is usually found. When allocation is not avoided but applied, the choice of energy or exergy allocation at the production stage is the most common choice. The use of other allocation approaches (mainly, mass and economic allocation) is found when multifunctionality appears before hydrogen production, namely at the stage of feedstock (key hydrogen carrier) production or energy carrier (driving energy) production. It should be noted that most of the case studies applying exergy allocation are presented in a single study (Neelis et al. [2004](#page-15-0)). Besides this study, exergy allocation is

applied only in one additional article (Wulf and Kaltschmitt [2013\)](#page-17-0). The latter represents also one of the few studies that carry out a sensitivity analysis on different allocation approaches.

Allocation at the hydrogen use stage is not generally required. It is found only in a case study addressing power generation with distinction between peak and off-peak electricity (Valente et al. [2015](#page-16-0)). On the other hand, the case studies with multifunctionality at the feedstock production level are based on first/second generation biomass (71 %) or microalgae (29 %). In these cases, the additional products are other marketable materials such as industrial wood or fodder (76 %), biofuels (19 %) and heat (5 %). Finally, the case studies with multi-functionality at the energy production stage are based mainly on non-renewable sources (63 %) rather than on renewable sources (37 %). In these cases, the additional products are electricity (33 %), marketable materials such as sulphur and carbon black (32 %), heat (28 %), or other fuels (7 %).

The general trend observed means a high level of agreement with FC-HyGuide recommendations. The hierarchy presented in ISO 14044 (ISO [2006b\)](#page-15-0), the ILCD Handbook (JRC [2010\)](#page-15-0) and the FC-HyGuide guidance document (Lozanovski et al. [2011\)](#page-15-0) is generally satisfied. Overall, system expansion is mainly applied (48 %), with allocation based on physical (39 %) and economic (13 %) relationships showing a lower presence.

3.2 Life cycle inventory analysis

The origin, as well as the quality, of the data necessary in the LCI phase affects significantly the reliability of the assessment. As underlined in the FC-HyGuide guidance document (Lozanovski et al. [2011](#page-15-0)), the selection of inputs to and outputs from the system shall be as representative as possible. This applies to the whole system at both foreground (processes under the influence of the central decision maker) and background (processes beyond the influence of the central decision maker) levels. Data Quality Rating (DQR) can be performed considering aspects such as data representativeness (technological, geographical and time-related), completeness, precision and consistency (JRC [2010](#page-15-0)). In this review, data quality is not addressed since in many publications, the authors disregard this aspect, thus preventing the identification of trends in data quality (trends other than the lack of information in this regard). Moreover, below 30 % of the publications include uncertainty analysis concerning relevant inventory data. In contrast, the information collected regarding data sources is appropriate for the evaluation of trends.

Figure [6](#page-10-0) shows the origin of the main data for each stage within the system's boundaries, with no distinctions between primary/secondary data. The label "real data" means direct measurements in plants on both industrial and laboratory scale as well as data coming from official statements for region- and

year-specific electricity/gas/waste mixes. The label "non-scientific literature" refers mainly to data from industrial datasheets and reports from non-scientific institutions. This type of data source plays a role mainly at the hydrogen use stage because data specification from manufacturers is usually available for PEMFC and vehicle inventories. Reports from research institutes and laboratories are considered as "scientific literature" along with published articles, handbooks and doctoral theses. Data from simulation and calculation are included under the same label. The storage stage shows a significant use of this type of data owing to calculations of energy for compression and liquefaction. Personal or confidential communications, rough assumptions and estimates are included in the category "other".

On the one hand, LCA databases (e.g. ecoinvent, GaBi and GREET model databases) and scientific literature are highly used for both foreground and background processes in order to fill data gaps from hydrogen production to distribution and use. Since hydrogen economy is not yet well established in the current context, there is a lack in primary data for foreground technological processes. The selection of database depends on several factors such as regional context and LCA software. The use of the GREET model (Wang [1996\)](#page-16-0) is partly responsible for the significant use of databases not only for background processes but also for foreground processes.

On the other hand, the use of real data and simulation/ calculation data is reduced and involves primary activity data for foreground processes. Figure 6 shows that real data are mainly available for processes related to energy carriers and feedstock production. In particular, the highest use of real data is found for the production of energy carriers due to the use of specific grid mixes in electrochemical case studies. Real data

Real data Simulation/calculation DLCA database Scientific literature DNon-scientific literature DOther SNot specified

Fig. 6 Use of data sources at different stages

found at the hydrogen production and donor production steps involve mainly measurements on the laboratory scale in biological case studies. Finally, it should be noted that a low number of case studies do not specify the stage affected by the data source or do not report clearly the origin of the data.

3.3 Life cycle impact assessment

Choices regarding the selection of life cycle indicators and LCIA methods for environmental characterisation are discussed in this section. Normalisation and weighting are not addressed since only 17 % of the studies consider these optional steps (studies using mainly Eco-indicator methods). Figure 7 shows the life cycle indicators most commonly selected by the authors (viz., global warming, acidification, energy consumption, eutrophication, ozone layer depletion, photochemical oxidant formation, abiotic depletion, human toxicity, land use and human health). This set of indicators covers 80 % of the total number of indicator occurrences in the publications reviewed. Since the choice of both life cycle indicators and LCIA methods is generally homogeneous for the different case studies within a publication, Fig. 7 is based on the choices by single publication. Figure 7 also shows the type of method used to quantify each indicator, distinguishing the family to which the method belongs but without distinction between versions. Even though this section is limited to environmental categories, it should be noted that only 15 % of the studies include also social and/or economic indicators in the analysis.

The total number of occurrences found for each indicator dashed line in Fig. 7—is provided by the secondary axis of the figure. Global warming (i1) is visibly the most common indicator addressed by the authors. Regarding this indicator, IPCC methods are the most common choice to characterise life cycle GHG emissions. Two types of study are considered under the "IPCC" label: studies using the single-issue IPCC method, and studies applying the IPCC characterisation factors to a limited set of emissions (typically, $CO₂$, N₂O and CH₄) without resorting to specific LCIA methods. Although the remaining LCIA methods usually apply IPCC characterisation factors to evaluate the global warming potential, they are classified separately in the "i1" bar of Fig. 7. A significant percentage of studies (ca. 10 %) do not specify clearly the method used to evaluate global warming.

The indicators on life cycle energy consumption are subdivided into the categories most commonly addressed in the studies: total energy (i3), fossil energy (i7) and nonrenewable energy (i8). Regarding these indicators, only studies addressing life cycle energy consumption for both foreground and background processes are considered in Fig. 7. Similarly to global warming, these indicators are significantly affected by studies that do not report explicitly the LCIA method applied. Concerning other indicators, it should be noted that "i11" includes only land use and land competition indicators (but not land use change). Finally, human health (i12) is found to be the most commonly assessed damage indicator, mainly using Eco-indicator methods.

Apart from the use of IPCC and VDI methods for the evaluation of global warming and cumulative energy demand (CED), respectively, CML is visibly the most commonly used family of methods. Even though some studies do not state clearly the specific version used, the CML 2001 version is found to be the most applied one. The use of CML methods is observed mainly in the European context. The general trends observed in Fig. 7 in terms of impact assessment methods are generally consistent with FC-HyGuide provisions, which recommend the use of CML impact methods (Lozanovski et al. [2011\)](#page-15-0). In terms of impact categories, FC-

Fig. 7 Choice of life cycle indicators and LCIA method. i1: global warming; i2: acidification; i3: energy consumption (total); i4: eutrophication; i5: ozone layer depletion; i6: photochemical oxidant formation; i7: energy consumption (fossil); i8: energy consumption (non-renewable); i9: abiotic depletion; i10: human toxicity; i11: land use; i12: human health (damage category)

HyGuide provisions recommend the use of global warming, acidification, eutrophication, photochemical ozone creation potential, primary energy demand (renewable and nonrenewable) and further categories such as land use and ozone depletion. These categories are in line with those most commonly found in the literature review.

LCIA results in terms of quantified impacts are not covered in this work. This is due not only to the focus on methodological choices but mainly to the inappropriateness of mixing results from a high number of LCA studies with different (and therefore inconsistent) methodological frameworks (Susmozas et al. [2015\)](#page-16-0). In this sense, an appropriate review of LCIA results requires the definition of a harmonisation protocol to provide consistent and comparable life cycle impacts that help identify the most favourable systems. The present review of methodological choices in LCA studies of hydrogen energy systems serves as a starting point for the definition and application of such a protocol for harmonised impacts, which is currently under development.

3.4 Agreement with FC-HyGuide provisions

Table [3](#page-13-0) summarises the level of accordance of the observed trends with a selected set of recommendations from the FC-HyGuide guidance document (Lozanovski et al. [2011](#page-15-0)). In this sense, the objective is not to address all FC-HyGuide provisions but to contextualise the trends identified in this review with the corresponding set of FC-HyGuide recommendations dealing with the same methodological issues. In Table [3,](#page-13-0) a very high level of agreement means that at least 90 % of the case studies are consistent with the FC-HyGuide recommendations for a given topic, while lower percentages mean high (60–90 %), intermediate (40–60 %), low (10–40 %) and very low $(\leq 10\%)$ level of agreement. Overall, the goal and scope definition and LCIA stages show the highest level of accordance, unlike the remaining stages (product system information and LCI).

When comparing the levels of agreement before and after the release of the FC-HyGuide guidance document, a slight favourable increase in providing technical details of the product system is generally seen, but not yet enough to conclude an overall satisfactory level of agreement regarding product system information. On the other hand, concerning goal and scope definition, a significantly satisfactory level of agreement is generally found both before and, especially, after FC-HyGuide, despite an opposite trend of agreement (from "very high" to "high") regarding the application of approaches to deal with multifunctional systems. In contrast, a favourable trend towards agreement is found for comparative studies regarding consistency in the methodological framework.

Concerning LCI, the lack of information on data quality and the limited use of primary data for foreground processes—issues that go against FC-HyGuide provisions—are seen both before and after the release of the FC-HyGuide guidance document. When filling data gaps with secondary data, an increase in the use of LCA databases rather than other sources is found when comparing the studies before and after FC-HyGuide. While information regarding data sources is usually provided (except for the specific version of the LCA databases used), a not negligible percentage of studies do not state the name and/or version of the LCA software used.

Finally, regarding LCIA, a general increase in the level of agreement between the trends observed and the corresponding FC-HyGuide provisions is observed when comparing the studies before and after FC-HyGuide. Nevertheless, while a satisfactory level of agreement is seen for the selection of the assessment method, a less satisfactory level is found for the selection of impact categories other than global warming. This is closely linked to the high number of studies addressing a reduced number of impact categories or even only one impact category (viz., global warming, due to the relevance of climate change in terms of sustainability concerns and energy policy).

4 Conclusions

Trends in methodological choices in LCA studies of hydrogen energy systems were identified and discussed through a complete literature review. Methodological and technological aspects were linked via subdivision of the hydrogen production processes in three technological categories: thermochemical, electrochemical and biological. The hydrogen production processes most commonly considered were found to be based on: steam reforming and gasification within the thermochemical category, electrolysis within the electrochemical category and fermentation within the biological category. Furthermore, second-generation biomass was found to be the most common "green" hydrogen donor in the thermochemical category, while solar and wind power were identified as the most common energy sources in the electrochemical category. When hydrogen use is included, PEMFC is the most common technology addressed both in mobility and stationary applications.

Regarding methodological choices in the goal and scope stage, a slight dominance of XtoGate over XtoGrave studies was detected. XtoGrave studies use hydrogen predominantly for mobility. In XtoGate case studies, the most common steps considered as the final gate within the system's boundaries are hydrogen purification and hydrogen distribution (to the refuelling station). The FU is commonly based on physical properties when addressing hydrogen production. In XtoGate case studies, the choice of FU is similarly distributed between mass and energy FU regardless of the specific technological category. The case studies addressing the use of hydrogen in mobility and power generation usually apply travelled distance and energy as FU, respectively. When dealing

Table 3 Agreement between trends observed and FC-HyGuide provisions

^a Level of agreement: very low, $\leq 10\%$ of the case studies; low, 10–40 %; intermediate, 40–60 %; high, 60–90 %; very high, $\geq 90\%$

^b Based on 46 publications (237 case studies) between the years 1998 and 2011

 \degree Based on 51 publications (272 case studies) between the years 2012 and 2015

^d Only for XtoGate case studies

^e Based on number of studies (instead of case studies)

with multi-functional systems (typically involving energy co-products such as heat and electricity), system expansion is commonly applied at the hydrogen-production stage. When allocation is applied, it is usually based on energy values. Nevertheless, when multi-functionality arises before the hydrogen-production step (i.e. at the feedstock or energy carrier production step), mass and economic allocation approaches, along with system expansion, are the most common choices.

Concerning methodological choices in the LCI stage, a general failure in providing information on data quality was found. On the other hand, information on data sources is usually reported. In this respect, a high use of data from LCA databases and scientific literature was found for both foreground and background processes through the different stages of the system.

Regarding methodological choices in the LCIA stage, global warming was the most addressed impact category,

followed distantly by acidification, eutrophication, ozone layer depletion and photochemical oxidant formation. Energy consumption is also widely addressed, but not always from an actual life cycle perspective. Besides the use of IPCC and VDI methods for the evaluation of global warming and energy demand, respectively, the most commonly applied methods belong to the CML family.

The evaluation of the level of agreement between the trends observed in this review and the recommendations given by the FC-HyGuide guidance document led to identify goal and scope definition and LCIA as the stages with the highest level of accordance, in contrast to the LCI stage. By comparing the studies before and after the release of the FC-HyGuide guidance document, a moving trend towards agreement with FC-HyGuide provisions was found mainly for product system information and LCIA choices. However, further efforts should still be made in future studies to provide information enhancing their traceability (e.g. stating clearly the key features of the additional products and providing results split by subsystem). In fact, lack of full traceability of the studies is seen as a key aspect affecting the articles reviewed (and LCA studies in general).

Regarding future research, the high level of detail attained in this review may facilitate its use as a valuable support to make more robust methodological choices in future LCA studies of hydrogen energy systems. Furthermore, this review constitutes a starting point for the harmonisation of the LCA results of hydrogen available so far in the literature, thus completing the set of information and trends provided in this work. Finally, further efforts should be made in order to move from LCA of hydrogen energy systems to LCSA of these systems.

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