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Decarbonization potential of fuel cell technologies in micro-cogeneration applications: spotlight on SOFCs in a Belgian case study

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Abstract

There is a plethora of fuel cell technologies, many of which hold great promise in terms of their decarbonization potential, which this paper aims to explore. In fact, this paper discusses the only two existing technologies on the market, Polymer Exchange Membrane Fuel Cells (PEMFCs) and Solid Oxide Fuel Cells (SOFCs). Unfortunately, these commercial systems mainly use natural gas as primary fuel due to its cost and practicality (easy transport and storage, existing infrastructures, etc.). Using Belgium as a case study, this paper shows that their GHG mitigation potential remains rather insignificant compared to the average individual carbon footprint if their fuel is not decarbonised. Even so, their mitigation potential would still be far from sufficient, and other measures, including behavioural changes, would still need to be implemented. Nevertheless, some emerging fuel cell technologies, such as Direct Carbon Solid Oxide Fuel Cells (DC-SOFCs) or Direct Formic Acid Fuel Cells (DFAFCs), offer the possibility of facilitating pure CO_2 capture at their anode outlet, thus allowing for potential negative emissions. Using a case study of the electricity demand of an average Belgian home (with two adults) supplied by an efficient biomass-fuelled DC-SOFC, this paper shows that these negative emissions could be up to about 4 tCO_{2eq}/year. By comparison, the IPCC's Sixth Assessment Report estimated the emissions footprint that could never be mitigated, even with future net-zero CO_2 emissions, to be 1 tCO_{2eq}/year per capita, implying that climate neutrality will require similar levels of carbon sequestration. In populous Western countries, natural carbon sinks are unlikely to be sufficient, and the potential negative emissions of emerging fuel cell technologies will be welcome.

Keywords: carbon budget; DCSOFC; fuel cell; negative emissions; CCU; CCS; carbon absorption

1. Introduction

In its Sixth Assessment Report (AR6), the Intergovernmental Panel on Climate Change (IPCC) established the remaining carbon budget that humanity can emit from 1 January 2020, with a 67% probability of limiting global warming to +2°C above pre-industrial levels, at 890 $GtCO_2$ [1].

One recognised difficulty is the allocation of this carbon budget among countries [2] (and individuals [3]). Two well-known approaches are the "grandfathering" principle (measures of "inertia") and the "equity" principle [4]. The "grandfathering" principle maintains that the carbon budget should be distributed among countries in accordance with their existing emission levels. In contrast, the "equity" principle asserts that the budget should be allocated in proportion to their population levels, i.e. that every human being has the same "right to pollute". The "grandfathering" principle is the subject of considerable criticism, primarily on the grounds that it favors "the perpetuation of an unfair allocation of rights on the basis of the previous unfair allocation of the same rights" [5]. This means that it is already unfair enough that historical per capita emissions were not equal across countries, so they should not be used as a basis for further (inequitable) carbon budget allocations. Consequently, if countries integrate carbon budgets into their climate strategies (as they should [6]), they usually adhere to the principle of "equity" [3]. This is exemplified by France and Wallonia [7], one of the three Belgian regions that serve as the case study in this paper.

As a final individual target, it is often considered that the 2050 carbon footprint should be capped between 1 and 2 tCO_{2eq}/year per capita [8]. For instance, a target of 2 tCO_{2eq}/year per capita is commonly adopted by online carbon footprint calculators [9]. However, it was recently underscored that, in order to achieve GHG (Greenhouse Gasses) neutrality in regions/nations such as France and Wallonia (or, by extension, Belgium), it is preferable to reduce the individual carbon footprint target to approximately 1 tCO_{2eq}/year per capita [3]. Indeed, that final GHG footprint will have to be absorbed territorially to meet the climate-neutral targets (such as those implied by the European Green Deal [10]). This could either be performed through nature-based, i.e. natural sinks [11], or technological methods [12]. However, the latter are still immature: they can be considered too risky [7] and ethically questionable [13] for climate policies to fully rely on them, although their development is still highly needed in the context of risk mitigation. Therefore, it has been demonstrated that, in densely populated regions/nations such as Belgium and France, reaching such a threshold of 1 tCO_{2eq}/year per capita with the capacities of natural carbon sinks will be challenging enough (to ensure GHG neutrality) [3]. In fact, the aforementioned study [3] posited that an increase in natural sinks of at least +300% above current levels would be necessary to achieve the desired outcome. This would entail maximising carbon uptake across all territorial areas through deep land use change considerations (implementation of intensive urban vegetation, alternative agricultural techniques, *etc*).

Moreover, at net-zero CO_2 emissions (assumed in 2050 for the case study selected in this study, based on the European Green Deal [10]), the IPCC has indicated that all GHG could not be completely mitigated as efficiently as CO_2. An 8 $GtCO_{2eq}$ yearly footprint would indeed remain for humanity [1]. In accordance with the “equity” principle and considering a projected 2050 global population of 7.735 billion people [14], this also leads to an individual unmitigated 2050 footprint of approximately 1 tCO_{2eq}/year per capita. Consequently; the 2050 final GHG footprint target will thus be represented solely by non-CO_2 GHG pollutants.

It is regrettable that current GHG emissions remain significantly above the target level. For illustrative purposes, consumption-based CO_2-only emissions (exclusive of the full carbon footprint) have been reported for the year 2021 at respectively 7.2 tCO_2/year and 7.0 tCO_2/year per capita for the European Union and China (excluding emissions from land use, land-use change, and forestry emissions) [15]. Individual carbon footprint figures (including all GHG) are however more difficult to aggregate. Nevertheless, the average individual carbon footprint for Europe was reported to be 9.0 tCO_{2eq}/year for the year 2015 [16], while for China it was 8.1 tCO_{2eq}/year for the year 2012 [17].

With regard to the selected case study of Belgium, the average individual carbon footprint has remained relatively stable over time and has been established to 16.5 tCO_{2eq}/year per capita for the year 2001 [18], 16 tCO_{2eq}/year per capita for the year 2007 [19], and more recently, to 15.4 tCO_{2eq}/year per capita for the year 2011 [20]. According to an open-access web application available since 2024 [21], which allows for visualizing countries’ and regions’ carbon footprints using Sankey diagrams and which is documented in detail in several publications [22–24], those order of magnitudes are confirmed. It is noteworthy that the same reference [21] reports a carbon footprint only slightly lower for the year 2019 (latest available data), i.e. of 15.2 CO_{2eq}/year per capita (considering a consumption-based accounting method).

At the residential scale, micro-cogeneration fuel cell technologies are regarded as a promising area of development, particularly in their three main markets, which are Japan, Europe, and South Korea, due the implementation of ad hoc-aimed subsidies and programs [25–27]. The approximate doubling of the market on an annual basis (at least up to around 2012) [26], is illustrated in Figure 1. It is notable that South Korea still has extremely ambitious targets for power generation fuel cells: 2.1 GW_{el} to achieve by 2040 for “fuel cell for domestic buildings” in general and 15 GW_{el} for utility fuel cell power plants, including exports. These figures represent a significant increase from the 7.1 and 370 MW_{el} reported in 2019, respectively [28–30].

Similarly, Japan has set an ambitious target of installing 5.3 million fuel cell micro-CHP (micro Combined Heat and Power) systems by 2030, which it hopes will provide heat and electricity to10% of Japanese homes [31]. At last, it has been reported that Europe aims to reach a capacity of 10 000 systems per year by 2021 [31,32], with the objective of achieving (and stabilising), in its “expected scenario”, a capacity of approximately 3 million micro-CHP installations in the residential sector in 2030 [33]. The majority of micro-CHPs would be represented by fuel cells, with the objective of meeting the “European Union Energy Goals 2030” [34]. This would correspond to a cumulative amount of approximately 15 million installations by 2030 [33].

In fact, the objective of this study is to quantify the current and expected carbon footprint mitigation potential of residential fuel cell micro-CHP systems in comparison to the average individual carbon footprint. Belgium is employed as a case study, utilising its average current dwelling as a representative example. A key objective of this research is to include in the study some emerging fuel cell technologies, such as Direct Carbon Solid Oxide Fuel Cells (DC-SOFCs) and Direct Formic Acid Fuel Cells (DFAFCs), that offer the capability of facilitating pure CO_2 capture at their anode exhaust, thereby potentially enabling negative emissions.

Indeed, technologies exhibiting high CO_2 purity streams represent a significant opportunity, particularly in light of recent reports indicating that more than 80% of the other technological carbon absorption projects currently fail [35]. This is particularly pertinent to the selected case study of Belgium (and neighbouring countries/regions), where the high individual carbon footprint is coupled with limited natural carbon sink capacities [3].

Consequently, this research makes a direct contribution to four Sustainable Development Goals (SDGs) [36]: sustainable and modern energy for all (SDG7), sustainable cities and communities (SDG11), sustainable production (and consumption) patterns (SDG12), and climate action (SDG13).

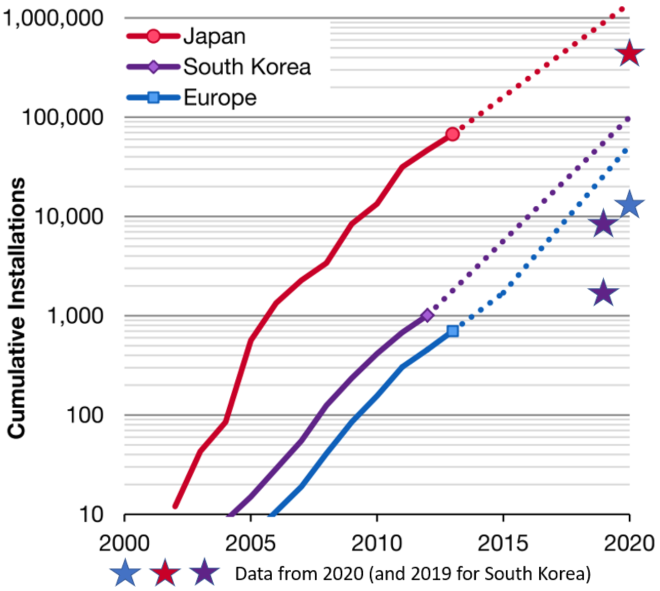

Figure 1. Cumulative number of residential fuel cell micro-CHP systems installed (solid lines) and near-term projections (dotted lines) reported in 2015. Reproduced and adapted from reference [37] with 2020 data for Japan and Europe [27] and 2019 data for Korea [29,30].

2. Materials and Methods

2.1. Review of fuel cell types

The number of existing fuel cell technologies and fuel cell acronyms is considerable. Such differences may be observed with regard to the electrolyte, the temperature of the stack, the fuel, *etc*. Even the same fuel cell acronym can be related to different fuel cell types. To illustrate Direct Carbon Fuel Cells (DCFCs), which utilise solid carbon as fuel, can exist with a variety of electrolytes, including oxide ceramics (DC-SOFC), alkaline solutions, or molten carbonate ceramics [38]. Each one of those DCFCs involves different electrochemical redox reactions, which constitute the core of the fuel cell working principle, thereby defining the reactants and their flows on the electrodes and through the electrolyte [38].

In light of the considerable diversity of emerging technologies, it has been observed that classifying fuel cells primarily according to their charge carrier more accurately represents the electrochemical redox reactions involved than the conventional classifications that rely on the fuel or the electrolyte [38]. In fact, the majority of fuel cell types can indeed be classified according to four categories of charge carriers : H^+ (protons), OH^-, O^{2-} and CO_3^{2-} [38].

Firstly, PEMFC (Proton Exchange Membrane Fuel Cell) [39], PAFC (Phosphoric Acid Fuel Cell) [39], MFC (Microbial Fuel Cell) [40], EFC (Enzymatic Fuel Cell) [41], the main type of DAFC (Direct Alcohol Fuel Cell) [42], DFAFC (Direct Formic Acid Fuel Cell) [43] and SOFC+ [44], also known as SOFC-H [45] (Solid Oxide Fuel Cells with Proton Conduction), SOFC(H+) [46], H-SOFC [47], PCFC (Protonic Ceramic Fuel Cell) [48], PC-SOFC [49], P-SOFC [50] or H+-SOFC [51], all utilise hydrogen ions (protons) as charge carriers, despite the differing nature of their fuel and electrolyte.

Secondly, the anion O^{2-} is employed primarily in Solid Oxide Fuel Cells with Oxygen-ion conduction (SOFC-O), but also in (SO-)DCFC (Solid Oxide Direct Carbon Fuel Cell, equivalent to DC-SOFC) [52]. Thirdly, MCFCs (Molten-Carbonate Fuel Cells) [43], also referred to as (MC-)DCFCs (Molten-Carbonate Direct Carbon Fuel Cells) [52], employ carbonate anions (CO_3^{2-}) as charge carriers. Lastly, (MH-)DCFCs (Molten-Hydroxide Direct Carbon Fuel Cells) [52] and AFCs (Alkaline Fuel Cells) [39], which encompass several fuel cell subtypes including the other main type of DAFCs (Direct Alcohol Fuel Cells) [42], utilise hydroxide anions (OH-) as charge carriers.

2.2. Available fuels for commercial fuel cell micro-CHPs

Regarding the fuel type, hydrocarbons, particularly natural gas, have become the most viable energy source for fuel cell micro-CHP systems, owing to their capability to be converted into hydrogen on-site (if necessary) [53]. For instance, in contrast to SOFCs for which the elevated operating temperatures of the stack may permit internal reforming [54], external reforming is required with PEMFC systems [55], which is generally incorporated into the embodiment of commercialised systems [56].

Natural gas is distinguished from other fuels by its cost-effectiveness and availability [57]. This is further evidenced by its substantial reserves, which, as of 2015, were projected to sustain (2015) extraction rates for 55 years [57]. Furthermore, its distribution network is well established in numerous regions globally [57]. It is the author's understanding that all commercially available fuel cell micro-CHP systems utilise natural gas, with the exception of some in Asia, where LPG (Liquefied Petroleum Gas) and kerosene have been reported to have been employed [57]. It is notable that some commercial systems are also reported as “hydrogen-ready”, particularly for applications where green hydrogen might be available [58].

2.3. Commercial micro-cogeneration fuel cell technologies

To date, only five fuel cell technologies have been considered for micro-CHP applications: Low and High Temperature Polymer Electrolyte Membrane Fuel Cells (LT-PEMFC and HT-PEMFC), Oxygen-ion Solid Oxide Fuel Cells (SOFC-O), Phosphoric Acid Fuel Cells (PAFC), and Alkaline Fuel Cells (AFC) [59,60]. Proton-conducting SOFCs (SOFC-H), which are typically considered as a subset of IT-SOFCs (Intermediate Temperature Solid Oxide Fuel Cells) [61], along with IT-SOFCs themselves, are still under study [38] and not regarded in this work as mature CHP technologies. Indeed, their electrolyte, anode, and cathode materials still need to be developed to achieve a sufficient power density and ensure the viability of their commercialisation [61]. Their commercialisation is in fact particularly limited by their cathode, which has traditionally exhibited poor activity or high thermal expansion [62]. Furthermore, SOFCs-H have been reported to have high manufacturing costs and scaling-up difficulties, which also impede their commercialisation [63].

Despite being developed earlier than PEMFCs and SOFCs, PAFCs and AFCs have not attracted substantial commercial interest due to the high manufacturing costs and low lifetimes respectively, inherent to these technologies [25]. Despite the absence of notable products that have been developed for the domestic CHP market utilising these technologies, they exhibit a multitude of desirable characteristics, at times even surpassing the performance of PEMFC CHP systems [64]. Previous studies have demonstrated or modelled these technologies at the 1-10 kW_{el} scale as CHP units, employing hydrogen directly as fuel [64,65]. It is noteworthy that PAFCs have been installed in large commercial and industrial applications for decades [27,66,67]. For instance, one commercial example has an electrical power output in the 100-400 kW_{el} range [66]. However, this is not the case for AFCs [27,67], likely due to their CO_2 intolerance and the necessity of a pure oxygen feed [38,68]. Nevertheless, CO_2 cost-effective removal apparatus could be conceived and has been developed [67]. In addition, similarly to LT-PEMFCs, there is a need to humidify the electrolyte through the inlet gasses for AFC technologies [69–71].

These limitations have been reported to predominantly confine PAFCs and AFCs to large-scale CHP applications (around 200 kW_{el}) and space vehicle uses, respectively [72], which is why these technologies are not further considered in this work for micro-CHP systems. In fact, to the best of the author’s knowledge, there is currently no HT-PEMFC system available on the market for residential micro-CHP applications. Furthermore, only a limited number of these systems have ever been (or are close to being) commercialised, as it has been reported in Table 1. For the sake of clarity, as indicated by their acronyms, it should be noted that HT-PEMFCs differ from LT-PEMFCs mainly through the higher temperature capability of their electrolyte [38].

Indeed, it has been recently reported that micro-CHP HT-PEMFC systems may only become commercially available in the near future, with their relatively short lifetime currently representing a significant obstacle to their widespread adoption [73]. In fact, a number of the remaining challenges that hinder HT-PEMFC commercialisation have been detailed in another recent publication, including the thermal instability of the catalyst [74].

Therefore, the only fuel cell technologies currently commercialised as micro-CHPs are (LT-)PEMFC and SOFC(-O), which have been compared in Table 2, especially in terms of LHV (Low Heating Value) electrical efficiency.

It is noteworthy that SOFC-PEMFC hybrid cogeneration systems have been studied approximately since 2000, initially based on the LT-PEMFC technology [75,76]. In such SOFC-PEMFC hybrid systems, owing to the internal reforming ability of the SOFC, this latter is used for both electricity generation and fuel reforming. Indeed, it can exhibit a reformate gas at its anode exhaust and subsequently fuel a downstream PEMFC, which allows for eliminating the need for a reformer upstream of the PEMFC [75,76]. A flowsheet of one example of an SOFC-PEMFC hybrid system is depicted in the following section in Figure 5. SOFC-PEMFC hybrid systems may be designed with the intention of combining some of the advantages of the two underlying fuel cell technologies, such as a higher energy efficiency (mainly by having removed the reformer of the PEMFC) while exhibiting a fast startup and a high stability [77]. However, until very recently, with the potential upcoming of the HT-PEMFC technology (if it ever becomes sufficiently mature, as inferred by Table 1), those hybrid systems have been reported impractical due to complicated post internal SOFC reforming components that are needed to reduce the CO levels at the fuel inlet conditions required by a LT-PEMFC [75]. Nevertheless, in a modelling study, the hybridisation of a SOFC and a HT-PEMFC at the micro-CHP scale has been recently investigated in 2024, showing promising results compared to standalone systems in terms of efficiency, emissions, and lifecycle cost [75].

2.4. Current and expected performance of micro-CHP systems based on a PEMFC or a SOFC

Back in 2010, the U.S. Department of Energy Hydrogen and Fuel Cells Program established the expected performance of micro-CHP systems in accordance with fuel cell types [78]. The maximum LHV electrical efficiencies reported attainable for LT-PEMFCs, HT-PEMFCs, and SOFCs(-O) fed by natural gas were, respectively, established at 40%, 45%, and 60% [78].

Actually, numerous studies have reported a maximum LHV electrical efficiency of approximately 45%for HT-PEMFC micro-CHP systems fed by natural gas, [37,73,78–80], thereby corroborating the aforementioned figure [78].

It is noteworthy that LT-PEMFC and SOFC(-O) commercial micro-CHP systems are already approaching those respective aforementioned efficiency figures [81,82].

Despite some studies [73,83,84] indicating that the LHV electrical efficiency of LT-PEMFC micro-CHPs could reach up to 45% (owing to significant technical improvements in the fuel reforming processor), it is usually assumed that it is probable that electrical LHV efficiency targets of 45% (or more) can be achieved with HT-PEMFCs and SOFCs, but unlikely to be achieved by LT-PEMFCs [37].

Table 1. List of HT-PEMFCs commercialised micro-CHP products and their current availability status.

HT-PEMFC CHP commercial attempts	Current status of the product or its manufacturing company
Enerfuel by Enerfuel [85–89]	Websites not available (even the one of the manufacturing company). It also seems that the Enerfuel company focuses on the transportation sector [86,89–91].
Elcore by Elcore [87,92,93]	The Elcore company declared bankruptcy in 2017 [94]. The Freudenberg company, which has taken over [94], stopped the commercialisation of HT-PEMFC CHP products to focus on fuel cell drive systems for heavy-duty vehicles [95] (probably because many flows have been reported on the Internet about the product [96]).
PureCell by ClearEdge [37,89,90]	The ClearEdge company declared bankruptcy in 2013 [97]. The Doosan company, which has taken over [97], seems to have stopped the commercialisation of the HT-PEMFC CHP products. They have mainly taken over the ClearEdge company for their industrial-scale Phosphoric Acid Fuel Cell (PAFC) catalog, such as the “PureCell Model 400” (400 kW$_{el}$) [98]. It was yet reported that HT-PEMFC micro-CHP systems are still under development at the Doosan company [99].
Gensys Blue by Plug Power [37,87,90,91,100–104]	Only field trials were operated and the product never made it to the market [91,100,103]. Indeed, Plug Power gave a “No-Go” on the project back in 2012 [100].
Serenus by Serenergy [90]	In fact, only fuel cell stack modules were developed. Indeed, they were not commercialised directly as CHP units and had to be integrated with other features, such as a fuel processor, since PEMFC systems require a high-purity hydrogen fuel [71] and since they are simply air cooled (no dedicated heat recovery feature) [91,105]. Furthermore, those fuel cell modules were mostly used in transportation applications [91]. It seems that the company (now named “Advent Technologies” [106]) no longer commercialises those systems but currently sells methanol-fed fuel cell systems with high temperature stacks. Those methanol fuel cells are DAFCs (Direct Alcohol Fuel Cells) [42] not designed for CHP but for transportation or backup power applications [89,107].

Similarly, both Europe and the International Energy Agency have set the 2030 LHV electrical efficiency target for micro-CHP fuel cells (fed by natural gas or biogas) at up to 65% [108,109]. This target seems to be achievable only with SOFC technologies [27,110–113]. It is worth mentioning that the “Bloom Energy” company, based in the U.S., has already been marketing a SOFC-based CHP unit, fed by natural gas (or biogas), since 2023 [114]. This unit has a power output of 330 kW_{el},which is higher than the micro-CHP range [59], and a net LHV electrical efficiency (AC) of 65% [114–116], which is higher than the efficiency targets set by the European Union and the International Energy Agency for 2030 [108,109].

In fact, the theoretical LHV electrical efficiency of an SOFC is close to 100% for the electrochemical direct oxidation of dry methane without consideration of parasitic losses, thus exhibiting no Carnot limit [117]. This is comparable to the findings reported for carbon-fuelled SOFCs (DC-SOFCs) [118]. With hydrogen or carbon monoxide, the theoretical efficiency of SOFCs is still approximately70% (LHV) [117]. This high 70% figure is, in fact, sometimes already reported as the current upper electrical efficiency limit for SOFCs with internal reforming [119,120].

In other recent developments within the (SOFC) industry, “Ceres Power” (United Kingdom) has announced a 65% LHV electrical efficiency for a 5 kW_{el} stack operating on methane (gross DC efficiency) [117,125]. Lately, the “Robert Bosch” company (Germany) has announced a 10 kW_{el} demonstrator with a gross LHV DC electrical efficiency of 70% [126]. Finally, both those companies have announced that they increased this gross DC efficiency to 72.4%, based on the LHV of methane, for a 150 W_{el} stack, developed within the framework of a collaboration [127]. Similarly, the “Elcogen” company (Estonia) is commercializing SOFC stacks for micro-CHP OEMs (Original Equipment Manufacturers) with an announced gross DC LHV electrical efficiency of 72% [128], which has also been reported at 74% in 2021 [115,129]. It is notable that it has been reported that a 74% DC LHV efficiency performance was also already achieved back in 2013 by the “SolidPower” company (founded in Italy and recently renamed “SolydEra”) [129,130].

Table 2. Comparison between the two sole commercial micro-CHP fuel cell technologies, i.e. the (LT-)PEMFC and the SOFC(-O) systems. Reproduced and adapted from references [43,131].

Fuel cell type & Charge carrier	**Typical electrolyte**	**Major contaminants**	**Stack operating temperature (°C)**	**Specific advantages**	**Specific disadvantages**	**LHV Electrical efficiency (%)**
PEMFC & H+	Solid Nafion®, a polymer	Carbon monoxide (CO) [a] Hydrogen sulfide (H_2S) [a]	60–80 Only low-temperature PEMFC are currently commercialised [121]	Highly modular for most applications High power density Compact structure Rapid startup due to low temperature operation Excellent dynamic response	Complex water and thermal management [a] Low-grade heat High sensitivity to contaminants [a] Expensive catalyst Expensive Nafion® membrane [122] Low fuel flexibility	40-60 (with H_2) Currently limited to 38.5 with CH_4 as some fuel needs to be burned to provide heat to a methane reformer [123]
SOFC & O^{2-}	Solid yttria-stabilised zirconia, i.e. YSZ, a ceramic	Sulfides	800-1000	High electrical efficiencies High-grade heat High tolerance to contaminants Possibility of internal reforming Fuel flexibility Inexpensive catalyst Simpler water management - SOFC can work at a perfect drying state [124]	Slow startup Low power density Strict material requirements High thermal stresses Sealing issues Durability issues High manufacturing costs	55-65 (with H_2) Currently limited to 60%-65% with CH_4 [114,121], i.e. still high thanks to the SOFC fuel flexibility

[a] Contaminants, thermal, and water management of PEMFC stacks have been discussed more deeply in another work [71].

It is noteworthy that this (still significant) discrepancy between gross DC and net AC efficiency may be attributed to the fact that DC gross electrical efficiency does not account for the parasitic power losses (in the current collection), the DC-AC power conversion losses, and the consumption of auxiliaries, such as the air blower power consumption (oxidizing agent supply at the cathode) [132]. Nevertheless, the aforementioned gross efficiency figures are so promising that it is not implausible to anticipate the advent of SOFC-based commercialised micro-CHP systems with LHV electrical efficiencies of 70%-80% or even higher in the foreseeable future. This is at least the assumption that will be considered in this work.

For information, for natural gas (and biogas) appliances (including fuel cells), HHV (High Heating Value) efficiencies can be obtained from the LHV efficiency figures thanks to the natural gas HHV to LHV ratio, which has been reported to be about 1.1094 [133], and which is extremely dependent of its chemical composition [134].

2.5. Negative emissions fuel cell technologies

It is this author's understanding that only two emerging fuel cell technologies offer the capability of facilitating pure CO_2 capture at their anode exhaust, thereby potentially enabling neutral (or even negative) CO_2 emissions [38].

Firstly, the DFAFC (Direct Formic Acid Fuel Cell), which is quite similar to an LT-PEMFC (especially in terms of electrolyte, charge carrier, contaminants, efficiency, or operating temperature [38]), can decompose the formic acid fuel into CO_2 and protons at the anode [135]. Its working principle is depicted in the bottom part of Figure 2, which presents a simplified (neutral) life cycle of CO_2 through an electroreduction unit and a DFAFC. This neutral CO_2 cycle, in which formic acid qualifies as an electrofuel, that is to say a liquid hydrocarbon derived from (renewable) electricity as the primary source of energy [136], has the advantage of being “short” as it does not rely on (slowly growing) biomass. The illustrated capture of CO_2, its conversion into formic acid, and the subsequent utilisation in the DFAFC constitute a CCU technology (Carbon Capture and Utilisation) [137]. It is noteworthy that formic acid fuel could be produced from biomass [135,138], which, if associated with CO_2 sequestration (of the off-anode gasses), could enable negative emissions (the electroreduction would thus not be needed). This would thus constitute a (BE)CCS technology, i.e. (Bioenergy) with Carton Capture and Storage [137].

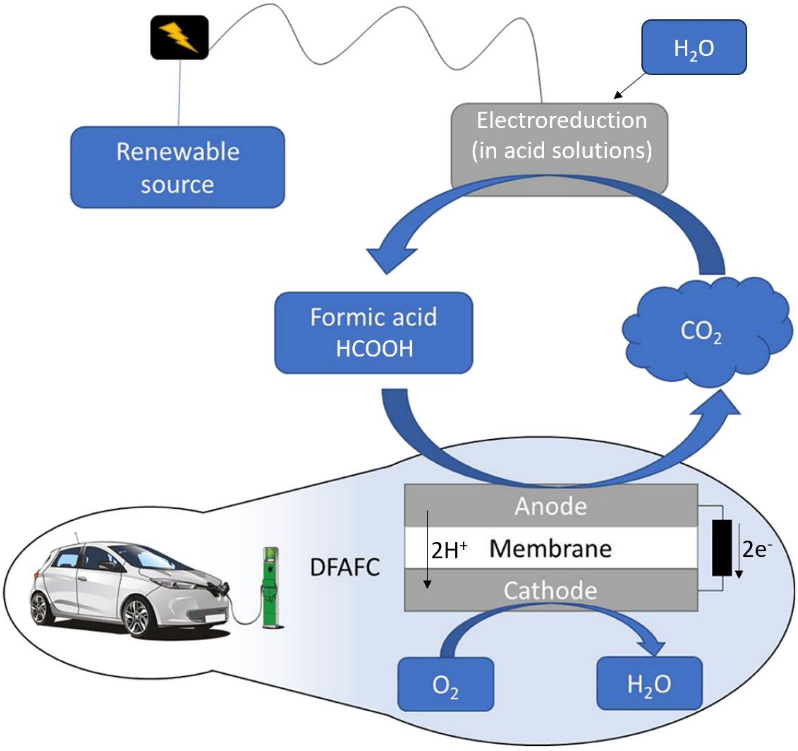

Figure 2. Simplified (neutral) “short” life cycle of CO_2 through an electroreduction unit and a DFAFC. Reproduced and adapted from references [135,139]. Copyright 2020 and 2023, American Chemical Society.

Secondly, the DC-SOFC (Direct Carbon Solid Oxide Fuel Cell), which is quite similar to a SOFC-O fed with hydrocarbons such as natural gas or biogas (especially in terms of electrolyte, charge carrier, sulfur contamination, efficiency, or operating temperature [38]) also exhibits an almost pure CO_2 stream at the anode exhaust [140]. Its working principle is depicted in the right part of Figure 3, which presents a simplified (neutral) life cycle of CO_2 through a methanation unit, a SOFC, and a (SO-)DCFC. The illustrated capture of CO_2, its conversion into biogas, and the subsequent utilisation in the

SOFC constitute a CCU technology (Carbon Capture and Utilisation) [137]. Negative emissions would trivially be obtained with a biomass-fed (SO-)DCFC if the high-purity CO_2 fuel cell exhaust stream was to be directly sequestrated instead of being converted into biogas, which would thus constitute a (BE)CCS technology, i.e. (Bioenergy) with Carton Capture and Storage [137].

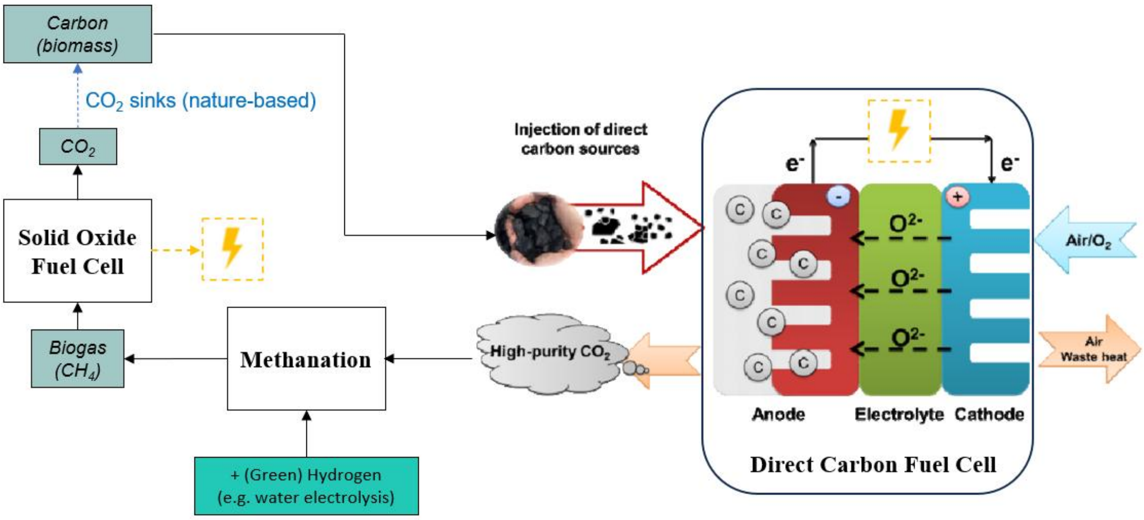

Figure 3. Simplified (neutral) "long" life cycle of CO_2 through a methanation unit, a SOFC, and a (SO-)DCFC. The DCFC working principle has been reproduced and adapted from reference [140] while the rest of the cycle has been established by the author of this paper.

This neutral CO_2 cycle has the disadvantage of being "long" as it relies on the slow growth of biomass. However, it offers the opportunity, for the same amount of biomass fuel, of combining the electrical production of two of the most efficient fuel cell technologies [38], which indeed exhibit theoretical electrochemical direct oxidation efficiencies that can be considered equal to 100% [117,118].

Similar other (CO_2-neutral) cycles involving CCU with (SO-)DCFCs have been reported in the literature, such as the one exhibited in Figure 4 [141], which illustrates a way of converting methane fuel into methanol and/or Dimethyl Ether (DME) while producing (green) electricity. Slightly different cycles have been reported by the same author in another paper [142]. In Figure 4, "Turquoise" hydrogen refers to hydrogen produced via pyrolysis of fossil fuels, where the buy-product is solid carbon (that can be stored or utilised, hence exhibiting a lower carbon footprint) [143]. A methanation unit could replace the CO_2 hydrogenation unit to produce methane instead of methanol and/or DME, allowing for completely closing the cycle and preventing the extraction of fossil methane. It is noteworthy that both methanol and DME are well-known as versatile alternative fuels potentially useful in many applications, such as for the transportation and the power sector [144].

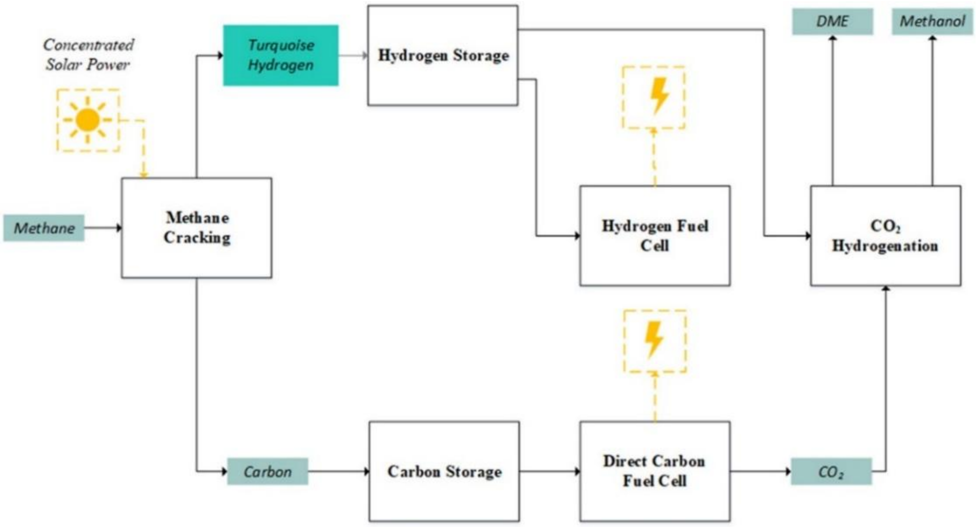

Figure 4. CO_2-neutral conversion of methane into methanol and/or Dimethyl Ether (DME) while providing (green) electricity, which constitutes a CCU technology via a (SO-)DCFC. Reused from reference [141].

Configuration possibilities based on Figure 3 and Figure 4 are endless but leave the scope of this research.

It should however be stated that, as mentioned in the paper used as reference and inferred by Figure 2 [135], DFAFCs are mainly attractive for small portable applications and promising for automotive applications (rather than for cogeneration). This is attributable to the intrinsic similarities between their characteristics and those of LT-PEMFCs. In fact, this is mainly due to their low electrical efficiency in conjunction with their high power density, their straightforward fuel storage, and convenient fuel delivery [38]. Actually, in light of the aforementioned lower anticipated efficiency figure of the PEMFC technology in comparison to (DC-)SOFCs, it has been reported that historical PEMFC manufacturers have reached the conclusion that the future of fuel cells in domestic built environment applications lies with SOFCs, and have therefore ceased PEMFC development [104]. Therefore, in this paper, lower efficiency fuel cell technologies such as the PEMFC and the DFAFC technologies they derive from will no longer be considered from here onwards to focus solely on (DC-)SOFC systems.

It is noteworthy that a SOFC-PEMFC hybrid system may also present negative emissions without complex sorbent-based carbon capture processes [145]. A typical biogas-fed SOFC can exhibit off-anode exhaust gasses containing several species that can be separated through well-known processes, such as CO_2, CO, H_2O (and, without off-anode recirculation, an excess of H_2) [38,146]. CO can be transformed into CO_2 and H_2 through a water gas shift reactor and if necessary, for the remaining CO, transformed into CO_2 with oxygen through a preferential oxidation catalyst [71,145]. The remaining H_2 is processed through a PEMFC (that contributes to the electrical production), leaving only the CO_2 with water (in a vapor state) [145]. It can be pointed out that using a HT-PEMFC instead of a LT-PEMFC would allow for not having to use the selective oxidation catalyst (and the supplied oxygen), as HT-PEMFCs are tolerant to the small quantity of CO that could remain in the flue gasses after the water gas shift reactor [38,71,75]. At the PEMFC anode exhaust, water can simply be liquified and separated from the CO_2 through several cooling and compression stages (through a carbon separation unit) [145]. The resulting CO_2 of high purity can then be liquified and sequestrated, to enable the negative CO_2 emissions potential of the whole system [145]. This SOFC-PEMFC hybrid system has been schematised in a CO_2-neutral cycle on Figure 5. The illustrated capture of CO_2, its conversion into biogas, and the subsequent utilisation in the SOFC constitute a CCU technology (Carbon Capture and Utilisation) [137]. Negative emissions would trivially be obtained if the high-purity CO_2 fuel cell exhaust stream of the carbon separation unit were to be directly sequestrated instead of being converted into biogas, which would thus constitute a CCS technology (Carton Capture and Storage) [137]. It is noteworthy that another way of separating H_2O, H_2 and CO_2 species effectively from a SOFC exhaust is through pressure swing adsorption processes [145].

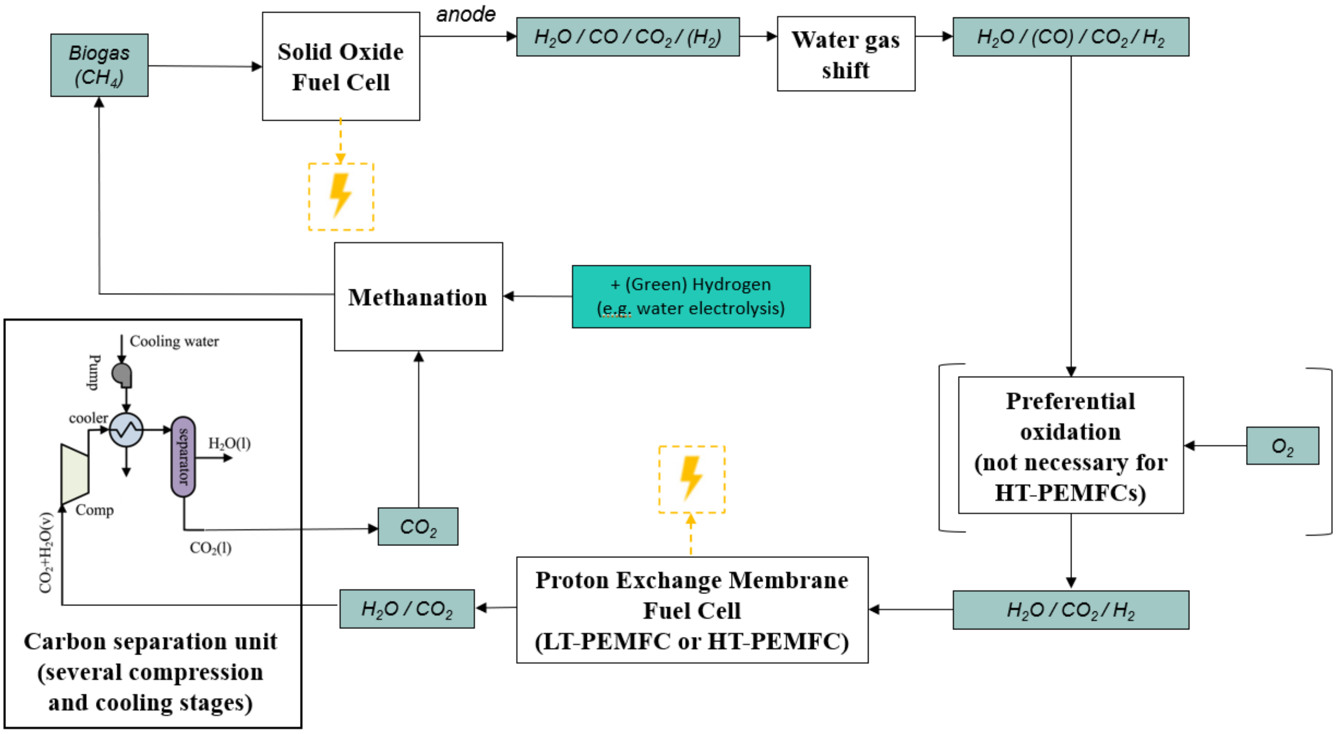

Figure 5. Simplified (neutral) cycle of CO_2 through a methanation unit, a SOFC, and a PEMFC. Simplified and adapted from reference [145].

2.6. Carbon footprint calculators

It has been chosen to use simple online carbon footprint calculators to illustrate the decarbonization potentials of the considered fuel cell micro-CHP technologies. In fact, for the purposes of demonstration, the selected individual carbon footprint calculator, which was first initiated back in 2014 [147], is that developed by AwAC, i.e. the Climate and Air Walloon Agency [148]. The primary reason for this selection is that it was developed by an official local agency. However, it should be noted that each carbon footprint calculator has its own specific accuracy, advantages, and limitations. Typically, the more complex and lengthy the associated survey is, the more accurate the results will be. However, this could present a significant barrier to accessibility for many individuals.

For example, the calculator in question does not take into account the impact of pet animals or of freshwater consumption. Similarly, the impact of "public services", i.e. the carbon footprint of collective public infrastructure and services (that, are equivalently shared for all citizens of a given country/region), is also not accounted for by the selected calculator. It is noteworthy that this "public services" footprint is the sole category that is not directly related to the individuals and that they are thus unable to directly influence. The responsibility for mitigating this category of emissions is therefore borne solely by the administration and its political choices.

It is therefore recommended that individuals utilise a number of carbon footprint calculators to better evaluate their sensitivity. Thus, for information and potential comparison purposes, a list of other well-known individual carbon footprint calculators has been provided in Appendix A.

It is of the utmost importance for users to be aware of the provenance of the calculators they are utilising, as the underlying assumptions may be contingent upon geographical factors (such as the emission factors of grid electricity, for example). Similarly, the tool should preferably have been recently created or updated. Also, even if most calculators express the carbon footprint per capita in terms of all-GHG emissions (CO_{2eq}), it is important to note that it is not always the case (some calculators may express their results in CO_2-only, for example).

It must also be remembered that such calculators only offer indicators and not accurate measured values. Indeed, the absolute value that results from those calculators is not really as important as its order of magnitude and trend over time.

In other words, the primary objective of such calculators is to provide a rough order of magnitudes of an individual's carbon footprint, and more importantly, to direct users towards the most impactful GHG mitigation actions they can implement at their individual levels. In fact, it is well-established that solving the climate crisis relies on changing human behaviour [149] and, to achieve it without redhibitory resistance, it is essential that applied policies and economics meet people where they are, utilising "audience-specific messaging and framing" [150]. Using individual carbon footprint targets, such as the 2050 1 tCO_{2eq}/year per capita considered in this paper, along with providing people with simple carbon footprint calculators for self-evaluation and action planning, is therefore highly pertinent in this context.

Even though methods of calculating carbon footprint are far from a global consensus [151], "raising awareness" by regularly monitoring one's carbon footprint is indeed often considered the first step towards behavioural change and carbon footprint mitigation actions [152]. As a similar example, it has indeed been demonstrated that energy motoring, performed regularly by the occupants of a dwelling, allows for a significant reduction in the energy consumption of that dwelling through behavioural change [153].

2.7. Individual carbon footprint example without dwelling energy uses

Individual consumption habits are always considered in carbon footprint calculators but they greatly differ from one person to another, and it is not within the scope of this paper to establish "average" habits of a given population. Furthermore, as the decarbonization potential of fuel cell micro-CHP systems is mainly associated with the dwelling's energy consumption, the assumptions pertaining to other domains of the individual carbon footprint are deemed inconsequential for the purposes of this study. Consequently, it has been decided to compute a single, realistic exemplar of an individual carbon footprint through the selected carbon footprint calculator. The assumptions implemented in the selected carbon footprint calculator are detailed in Appendix B for the purpose of reproduction.

Those assumptions may still be regarded as representative of the typical case, in the sense that they are not describing extremely unrepresentative cases, such as a fully vegan diet coupled with only light-mobility uses or a fully red-meat diet accompanied by extensive travel in a heavy diesel vehicle.

The resulting carbon footprint simulation example (excluding energy use of the dwelling) is shown in Figure 6 and is approximately 10 tCO_{2eq}/year—significantly above the 2050 target of 1 tCO_{2eq}/year per capita required for climate neutrality (based upon the unmitigable GHG emissions reported by the IPCC [1] and projected global population [14] considered in this study). This illustrates that even if dwellings achieve zero emissions from energy use, changes in behaviour and consumption choices remain essential.

Although it has been mentioned that the related assumptions (detailed in Appendix B) were not intended to represent the case of the average Belgian or European citizen, the resulting carbon footprint reported in Figure 6 (10 tCO_{2eq}/year per capita without dwelling energy uses) is consistent with the 15.2 tCO_{2eq}/year per capita (complete footprint) established for Belgium for the year 2019 [21–24].

It is worth mentioning that the remaining CO_{2eq} emissions still reported for the dwelling in Figure 6 are mostly related to the GHG emissions embodied in its construction (and that are vented onto the building's operational lifespan).

Up to this point, no consideration has been given to the energy uses of the dwelling (gas/electricity consumption). Consequently, the resulting individual carbon footprint (of 10 tCO_{2eq}/year per capita for the example given in Figure 6), is not related to the choice of the space heating appliance or the utilisation of any cogeneration system, whether fuel cell-based or otherwise.

In other words, it can already be stated with certainty that implementing fuel cell micro-CHPs, even when operating at 100% efficiency and fed by 100% biogas (or other climate-neutral fuels) represents an insufficient solution. Further action is required, including behavioural and consumption choice changes. It can also be acknowledged that negative emissions technologies, such as DC-SOFC micro-CHP systems, which are the subject of this investigation, can also provide significant benefits in this regard.

2.8. Individual carbon footprint example with energy uses

Additionally, this paper employs the average Belgian dwelling as a case study to illustrate the decarbonization potentials of fuel cell micro-CHP technologies. According to the Belgian regulator [154], it corresponds in 2023 to 17 000 kWh of natural gas consumption and 3500 kWh_{el} of electrical consumption per year. Those figures have been reported in Table 3.

Emission factors for gas and electricity consumption (mainly applicable to Belgium) have been extensively documented in a prior study [155]. Two LCA (Life Cycle Assessment) emission factor datasets, namely the dataset "A" and the dataset "D1" from the aforementioned research [155] have been replicated in Table 3 and employed in this study.

Dataset "A" was initially provided for the purpose of establishing CO_{2eq} savings from CHP units when compared to "reference systems", which consist of a gas condensing boiler of 90% of average LHV efficiency for heat production and in a CCGT (Combined Cycle Gas Turbine) of 55% of average LHV efficiency for electrical consumption [156]. This is not only relevant to Belgium but also to all regions/nations that regularly utilise CCGTs on their grids. It basically assumes that the decentralised CHP electrical production (e.g. of fuel cells) is replacing the electrical production of CCGTs not equipped with heat recuperation, which would generally require a district heating connection due to the size of the CCGT plants [157]. While this is a promising approach in the context of the energy transition, current CCGTs are not generally connected to district heating networks [158].

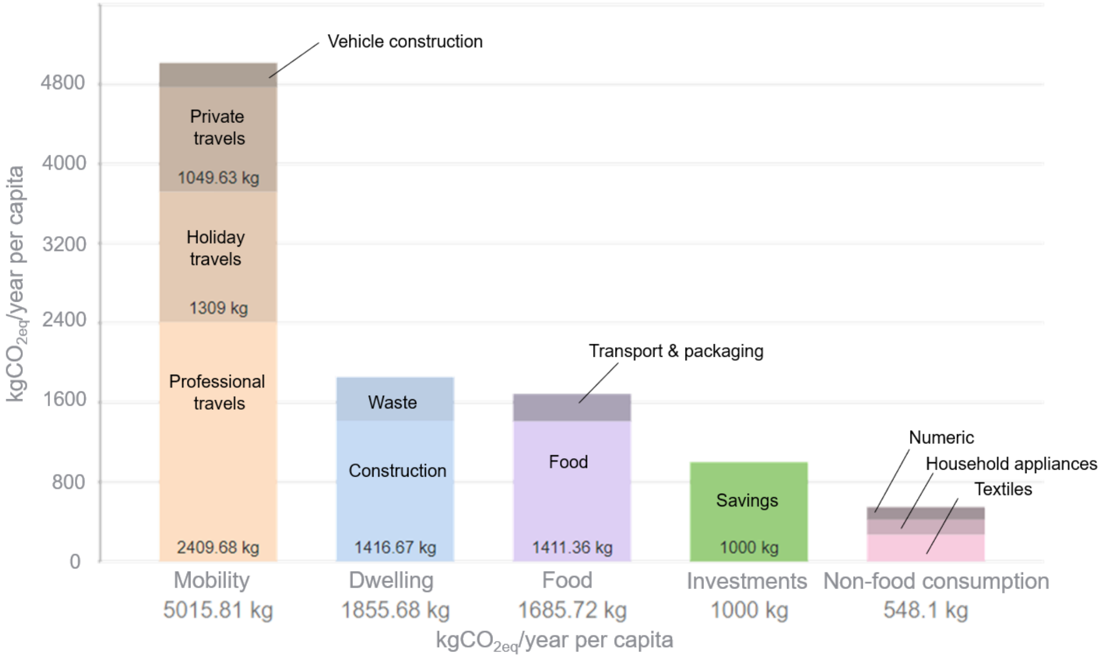

Figure 6. Chosen example of individual carbon footprint, without any energy uses considered for the dwelling (gas/electricity consumptions).

Dataset “D1” considers the Belgian statistical average electrical consumption emission factor between the population of unweighted hourly emission factors provided by Electricity Maps [159] for the whole year 2021. It therefore better corresponds to the average (Belgian) electrical mix than the dataset “A”.

However, none of the considered emission factors (neither dataset “A” nor dataset “D1”) take into account the electrical transportation and distribution losses (which can reach about 6-7% in the European Union [160]) that are avoided with decentralised electrical production systems, such as (micro-)CHP.

Table 3. Chosen emission factors (from a previous study [155]) for the energy uses of the average Belgian residential dwelling, which is represented by its current average energy demands (reported in 2023) [154]. The subsequent carbon footprints of the average Belgian dwelling’s energy uses are also established accordingly.

Dataset	Gas consumption emission factor (gCO_{2eq}/kWh)	Electricity consumption emission factor (gCO_{2eq}/kWh_{el})	Average gas consumption of the dwelling (kWh/year)	Average electricity consumption of the dwelling (kWh_{el}/year)	Carbon footprint (tCO_{2eq}/year)
Dataset “A”	251	456	17 000	3500	5,86
Dataset “E1”	254	167	17 000	3500	4,90

Actually, the electrical market and electrical prices (at least in the European Union) rely on the System Marginal Price (SMP) [161], which signifies that they are defined by the latest power plants that must be activated to meet the demand. These have typically been and, in Belgium, continue to be CCGT plants, either for reasons of flexibility and/or economic benefit. In fact, according to the hourly data provided by Electricity Maps [159] for the year 2021 (that have been used to compute the dataset “D1”), there was always some electrical production originating from natural gas power plants. As again evidenced by the data provided by Electricity Maps [159], this was also the case for the year 2020. Therefore, through the SMP principle, the Marginal Emission Factor (MEF), which “reflects the emissions intensities of the marginal generators in the system, i.e. the last generators needed to meet demand at a given time” [162], can be regarded as consistently equivalent to the emission factor for electricity production from natural gas power plants, as reported by the dataset “A” in Table 3. Furthermore, this is anticipated to remain the case in Belgium for the foreseeable future, considering the fact that the government is currently supporting the construction of new CCGTs, which are intended to (at least partially) phase out old nuclear power plants [163,164].

For this study, and at least for Belgium, both dataset “A” (marginal emissions) and dataset “D1” (average emissions) from Table 3 can thus indeed be considered relevant, with dataset “D1” holding even greater significance than dataset “A.”.

The average Belgian dwelling energy consumptions related to datasets “A” and “D1” respectively correspond to carbon footprints (of energy usage) of 5.86 tCO_{2eq}/year and 4.90 tCO_{2eq}/year, which must be be added, in proportion to the number of occupants in the dwelling, to the individual carbon footprint established in Figure 6 (which was close to 10 tCO_{2eq}/year). When divided by the number of adult occupants considered (i.e. two, as reported in Appendix B), those energy uses carbon footprint figures account for only approximately 25% of the total individual carbon footprint chosen example. This example once again emphasises the necessity for carbon footprint mitigation to extend beyond dwelling energy uses.

These constitute from here onwards the “base scenario” of this research, which will be used as a reference to illustrate the decarbonization potential of fuel cell micro-CHP technologies. To sum up, regardless of the emission factor dataset from Table 3 (either “A” or “D1”), the reference “base scenario” does not include any (micro-)CHP system and consistently assumes that the dwelling’s heat demands are supplied with a gas condensing boiler (of 90% of LHV efficiency). Additionally, it assumes that the electrical demand is supplied by the grid, with the corresponding assumption regarding the electrical emission factor.

3. Results

As it has been demonstrated that other fuel cell technologies are either not mature enough and/or not as efficient, this study has been focusing on SOFC technologies, which specific advantages have been presented in Table 2, and on DC-SOFCs they derive from. Consequently, Table 4 evaluates the carbon footprint of several “ideal” (DC-)SOFC micro-CHP systems that would correspond to an electrical production that would match the electrical consumption of the average Belgian dwelling given in Table 3 (3500 kWh_{el}). It also establishes the carbon footprint savings permitted by those fuel cells in comparison to the “base scenario” established in the preceding section. The thermal demand is assumed to always be equal to the 2023 average Belgian dwelling given in Table 3 (corresponding to a 17 000 kWh of gas consumption with a gas condensing boiler averaging 90% of LHV efficiency). In all cases, it is assumed that the implementation of the fuel cell micro-CHP will reduce the thermal demand for the gas-condensing boiler. As the energy transition progresses, it is likely that the renovation of buildings will result in a decrease in the thermal demand of the average dwelling. Conversely, the electrification of mobility is likely to result in an increase in the electrical demand of the average dwelling (in comparison to the demand considered in this study) [165]. This is however not taken into account at this point.

For the sake of simplicity, all the "ideal" cases considered in Table 4 assume that the (DC-)SOFC is fully flexible in terms of electrical production, matching the dwelling's electrical load profile exactly. It is not yet realistic to expect that current SOFC systems will be able to be shut down (or modulated down to 0%) due to their long startup times [54]. Nevertheless, the modulation range of SOFC systems currently on the market is already quite large (in the 33%-100% range [166]) and it is reasonable to hope that it will even be extended in the future.

The efficiencies of the fuel cell systems reported in Table 4 are based on the already discussed current and expected performance of the technology, as indicated here below :

- First case (1): 60% and 25% LHV electrical and thermal efficiency for the current performance of commercialised micro-CHP SOFC systems. It is noteworthy that these performances have been stated by the OEM [166] but have been verified in laboratory investigations [54,167] and, at least for the electrical efficiency, in field-test monitoring studies [166].
- Second case (2): 75% of expected LHV electrical efficiency for the SOFC technology seems realistic, as inferred in the previous section. It has however been assumed that this increased electrical efficiency would result in a five-percentage-point reduction in the thermal efficiency, thus maintaining the total LHV efficiency at a realistic level as well of 95%.
- Third case (3): same SOFC as in the second case but entirely fed by biogas (with an assumed emission factor of zero). The remaining heat demand of the dwelling is also assumed to be provided with "green energy" (with an assumed emission factor of zero).
- Fourth case (4): similar assumptions as in the third case, with a SOFC exhibiting slightly increased performance. In this instance, the SOFC is however fed with solid biomass, i.e. dry biochar such as pinewood biochar (also with an assumed emission factor of zero). It would therefore be a DC-SOFC which, as previously discussed, produces a highly pure CO_2 stream at the anode exhaust, thereby enabling the capability of CO_2 capture (and negative emissions). It should however be mentioned that the maturity of DC-SOFC is currently insufficient [43] to expect the introduction of such systems within the short timeframe required for the energy transition [3]. Nevertheless, its potential for negative emissions may still prove beneficial in the future, regardless of the timing of the emergence of the technology, as it will be demonstrated subsequently in this section. Although the theoretical DC LHV efficiency of the electrochemical oxidation of solid carbon is close to 100% [118], 80% LHV electrical efficiency for real DC-SOFC micro-CHP systems seems to be a more realistic efficiency figure, which is also often considered in the reviewed literature [168]. It has again been assumed that this increased electrical efficiency (in comparison to the third case) would result in a further reduction of the thermal efficiency by five percentage points, in order to maintain the total LHV efficiency at a realistic level of 95%.

Thus, compared to average grid electricity, Table 4 demonstrates that the impact of fuel cells fed by natural gas (or other fossil fuels) is detrimental when compared to the "base scenario", as evidenced by the negative savings reported in Table 4. Even when a comparison is made with the electrical production of CCGTs with a high emission factor of 456 gCO_{2eq}/kWh_{el} (marginal emissions), and when highly efficient future natural gas flexible SOFCs are taken into account (second case), Table 4 only indicates small carbon footprint savings in comparison with the "base scenario" (positive savings reported in Table 4). In view of the desirable reduction in the use of CCGTs, this once again emphasises the importance of flexible electrical production for future fuel cell micro-CHP systems (at least if they are not fed with CO_2 neutral fuels). Fortunately, this is already being achieved to a certain extent with some micro-CHP SOFC systems that are currently available on the market [166].

It is in fact only the introduction of 100% biogas (or other CO_2 neutral fuels), as considered in the third case, that exhibits the most substantial decarbonization potential. Indeed, as previously stated, the implementation of any (SOFC) micro-CHP systems, regardless of their efficiency (as in the first and second cases), results in a relatively insignificant or even negative CO_2 impact.

It is noteworthy that the carbon footprint savings reported in Table 4 are established in comparison to the reference "base scenario", considering current values of natural gas and electricity emissions factors. They have not been established in comparison to future potential reference scenarios involving greener grid gas and greener grid electricity. The objective is to evaluate the potential carbon footprint savings in comparison to the existing situation, i.e. to the reference "base scenario". It should however be noted that if CO_2 neutral fuels were to be available in the future for SOFCs, it is likely that they will also be used in other space heating and/or electrical production appliances. For example, biogas can currently be readily employed in a gas condensing boiler (or even in a CCGT power plant). Consequently, the genuine decarbonization potential of micro-CHP systems can be attributed to negative emissions technologies, such as DC-SOFCs (as evidenced in the fourth case of Table 4).

Table 4. Yearly carbon footprint calculations of several SOFC micro-CHP systems energy use corresponding to an average Belgian dwelling of 17 000 kWh and 3500 kWh_{el} of gas and electricity consumption.

Data	Best "current SOFC", 60% LHV electrical efficiency, 25% LHV thermal efficiency (1) [a]	Best "future SOFC", 75% LHV electrical efficiency, 20% LHV thermal efficiency (2) [a]	Best "future SOFC", 75% LHV electrical efficiency, 20% LHV thermal efficiency, 100% biogas (3) [a]	Best "future DC-SOFC", 80% LHV electrical efficiency, 15% LHV thermal efficiency, 100% biochar with CO_2 capture (4) [a]
Fuel cell electrical production (kWh_{el}/year)	3500	3500	3500	3500
Gas consumption related to the fuel cell electrical production (kWh/year)	5833	4667	4667	0
Fuel cell heat production (kWh_{th}/year)	1458	933	933	656
Remaining heat demand, supposably provided by a 90% LHV efficient gas boiler (kWh_{th}/year)	13842	14367	14367	14644
Total (fossil fuel) gas consumption, fuel cell, and gas boiler (kWh/year)	21213	20630	0	0
Carbon footprint related to the (fossil fuel) gas consumption - Dataset "A" (tCO_{2eq}/year) - marginal emissions	5,32	5,18	0,00	0,00
Carbon footprint related to the (fossil fuel) gas consumption - Dataset "E1" (tCO_{2eq}/year)	5,39	5,24	0,00	0,00
Negative carbon footprint from CO_2 capture at the anode exhaust [b]	N/A	N/A	N/A	1,76
Carbon footprint savings - Dataset "A" (tCO_{2eq}/year) - marginal emissions	0,54	0,68	5,86	7,63
Carbon footprint savings - Dataset "E1" (tCO_{2eq}/year)	-0,49	-0,34	4,90	6,67

[a] The different cases are based on the current and expected/hoped performance of fuel cell micro-CHPs. Those calculations are related to an average dwelling and are not expressed as "per capita". All fuel cells are assumed to be fully flexible to meet the dwelling's electrical demand.
[b] With the carbon content intensity assumption equivalent to 403 gCO_2/kWh_{fuel} (resulting figures are expressed in the table in tCO_2/year or tCO_{2eq}/year in this case, as CO_2 can be assumed to be the only GHG released fuel cells [133]). This carbon content intensity has been calculated for dry pinewood biochar (HHV = LHV = 24.49 MJ/kg, 59,86% of carbon content, DC-SOFC with 80% of electrical efficiency [168]). It is crucial to consider that detrimental positive GHG emissions related to biochar production (and transport) have been neglected for the sake of simplicity and to be consistent with a low-carbon economic future (where, for example, the GHG emissions of the transportation and energy sectors can be neglected and where biochar can grow without GHG-emitting fertiliser). If those detrimental emissions were to be considered, this would straightforwardly tend to reduce the negative emissions potential of the considered DC-SOFC reported in this table. Life cycle emissions of the fuel cell itself have also been neglected because of a lack of available data (construction, transport, disposal, *etc*).

The carbon footprint savings due to the implementation of the ideal fuel cell micro-CHP systems in the average Belgian dwelling outlined in Table 4 have been reproduced graphically in Figure 7. This has been performed on the assumption that the dwelling in question is occupied by two adult individuals, as detailed in Appendix B. Actually, in an average Belgian dwelling such as the one represented by the energy consumptions considered in this section, there might also be some children. However, it has been decided to report the established carbon footprint of the dwelling solely on its (two) adult occupants. The average Belgian individual carbon footprint being close to one of Wallonia, i.e. about 15.4 tCO_{2eq}/year per capita [20], as it is one of the three Belgian regions, those decarbonization potentials have been represented over two Walloon individual carbon footprint pathways compatible with IPCC's +2°C "equity" carbon budget established in a previous study [3]. Indeed, it can be seen in both scenarios that the blue bars, corresponding to IPCC's remaining +2°C "equity" carbon budget, represent positive values in the year 2050. The GHG mitigation scenario illustrated on the left even never allows for the carbon budget to be exceeded (and reach negative values), as opposed to the right one. Only the decarbonization potentials established from marginal emissions (and reported in Table 4) have been considered (i.e. using dataset "A" from Table 3). Indeed, dataset "D1" (in Table 3) has not been used because there are no carbon footprint savings for natural gas-fed fuel cells when average grid electricity is considered (as exhibited in Table 4). The "O" dots correspond to the reference "base scenario", i.e. the Belgian average dwelling energy uses carbon footprint given in Table 3 (using the marginal emissions provided by dataset "A"). All of the dots have been established by dividing the Belgian average dwelling energy uses carbon footprints (deduced from or directly reported in Table 3 and Table 4) by the number of adult occupants in the dwelling (i.e. two in this case, as stated in Appendix B), and added to the chosen example of carbon footprint without energy uses reported in Figure 6. It can again be deduced that significant decarbonization can only occur using CO_2 neutral energy vectors (dots "3"), not necessarily in fuel cell systems. However, a significant decarbonization potential of fuel cell systems still lies in technologies that exhibit a highly pure CO_2 stream at their exhaust and thereby facilitate CO_2 capture, such as DC-SOFCs (dots "4").

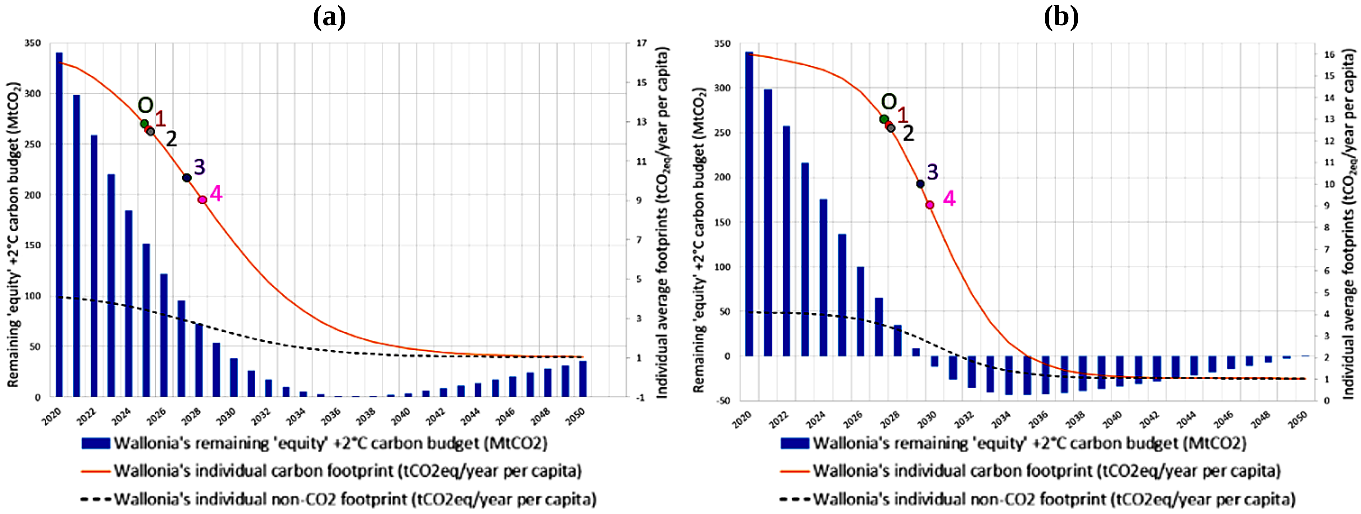

Figure 7. Decarbonization potentials of the implementation of the four "ideal" cases of (DC-)SOFC micro-CHP systems reported in Table 4 for an average Belgian dwelling (dots "1", "2", "3" and "4"). Reproduced and adapted from reference [3].

The initial carbon footprint of the reference "base scenario" (dot "O" in Figure 7, i.e. about 13 tCO_{2eq}/year per capita) is actually lower than the current average individual carbon footprint in Belgium. As observable in Figure 7, the individual carbon footprint of Wallonia (or by extension, that of Belgium) shall actually reach this value at least between 2025 and 2028, depending on the mitigation scenario, i.e. Figure 7(a) or Figure 7(b). In other words, the assumptions set out in Appendix B and implemented to the selected carbon footprint calculator (which produced Figure 6), along with the energy use assumptions (per capita) related to the average Belgian dwelling (reported in the previous section) could actually be regarded as a few years ahead of the current average Belgian individual carbon footprint, in light of the proposed GHG mitigation pathways illustrated in Figure 7.

4. Discussion

As evidenced in Table 4, the DC-SOFC exhibits a negative CO_2 emissions potential of 1,76 tCO_{2eq}/year for the considered current average Belgian dwelling. Although not insignificant, it may appear relatively modest in comparison to the current individual Belgian carbon footprint, which is estimated to be 15.2 tCO_{2eq}/year per capita for the year 2019 [21–24]. This is particularly the case, as illustrated in Figure 7, when the savings are divided by the number of (adult) occupants in the dwelling, which is in this instance two. Furthermore, it would even be reduced if this study were to consider the detrimental positive GHG emissions related to the fuel production (and transport), i.e. biochar in the case of the considered DC-SOFC, in addition to the negative emissions potential of CO_2 at the fuel cell exhaust. Indeed, these emissions were excluded in order to align with the assumption of a low-carbon economic future, as documented in Table 4. It is noteworthy that, as previously stated, the life cycle emissions of the fuel cell itself have also been excluded due to a lack of available data (construction, transportation, disposal, *etc*), which would further reduce this negative emissions potential.

Nevertheless, it has already been mentioned that the electrification of mobility (and of other areas of usage) is likely to result in an increase in the average electricity demand of the average dwelling. Therefore, the electrical demand (to the DC-SOFC reported by the fourth case of Table 4) will likely greatly exceed the 3500 kWh_{el} considered here, thus increasing the potential for negative emissions. To illustrate, a 20 kWh_{el}/100 km electric vehicle with 20 000 km annual mileage would necessitate an additional fuel cell demand of 4000 kWh_{el}. In addition to the initial domestic demand of 3500 kWh_{el}, this would result in an annual electric demand of 7500 kWh_{el} for the fuel cell. Considering the DC-SOFC of the fourth case of Table 4 (with 80% of LHV electrical efficiency), negative emissions could reach values slightly lower than 4 tCO_2/year for the dwelling (i.e. about 3.8 tCO_2/year). This equates to approximately 2 tCO_2/year per capita when considering the number of adult occupants in the household. Alternatively, when the current average fertility rate in Europe of 1.6 [169] is also considered (which increases the considered number of occupants of the dwelling to 3.6), the figure rises slightly above than 1 tCO_{2eq}/year per capita. Actually, in recent studies for developed countries [170], the annual distance travelled with a passenger car is commonly assumed to be 16 000 km (in contrast to the 20 000 km considered here). However, this has a negligible impact on the results as, when the 1.6 current average fertility rate in Europe [169] is considered (i.e. 3.6 occupants in the dwelling), the negative CO_2 emissions potential of the DC-SOFC micro-CHP is only reduced to a value slightly below 1 tCO_{2eq}/year per capita. This approach can be intensively criticised on the grounds that it suggests that the greater electrical consumption of the dwelling, the greater the negative emission potential of the DC-SOFC associated with it. While this has indeed been demonstrated mathematically, it runs dangerously counter to the “energy sobriety principle”, sometimes known as “enoughness” or “frugality”, but better known as “sufficiency” [171], as introduced in the IPCC’s Sixth Assessment Report in 2022 [172]. Whatever the semantics, sufficiency[1] should always be the primary and necessary consideration in the energy transition [172–174]. In addition, it seems highly unlikely that a significant deployment of DC-SOFCs without sufficient measures to promote sufficiency/sobriety would be without considerable risks. For instance, critical concerns could indeed be linked to tensions around material or biomass availability or to various environmental issues (such as unexpected pollution levels and/or harmful impacts on biodiversity), which could exert pressure on the defined limits of planetary boundaries [175].

Nevertheless, the potential negative emissions allowed by the electricity production from biomass-fed DC-SOFCs can still easily be even higher than the recommended 2050 individual carbon footprint (reduced from current levels, considering the sufficiency/sobriety principle as the first mitigation step). It has indeed been established that the latter is also estimated around 1 tCO_{2eq}/year per capita, based on the already discussed unmitigable GHG emissions reported by the IPCC [1] and the projected global population [14]. It is noteworthy that the achievement of full climate neutrality implies that (natural or technological) carbon absorption will also reach this 1 tCO_{2eq}/year per capita target by 2050. Fortunately, it has been shown here that the potential for CO_2 sequestration at the anode exhaust of biomass-fuelled DC-SOFC micro-CHPs in residential applications could be in this order of magnitude.

Actually, technical negative emissions technologies, examplified by DC-SOFCs, will assume a pivotal role in the future if the natural territorial absorption levels are unable to be sufficiently increased. Indeed, as previously discussed, attaining such a level of carbon absorption with natural sinks alone will prove to be a significant challenge [176], especially for densely populated regions. According to a recent study [3], a minimum increase of +300% in carbon sinks would be necessary for regions/nations like Belgium and France compared to their current levels.

It is important to note that these potential negative emissions would be additive to the trivial benefits obtained from the avoided fossil fuels permitted by electrification of societal uses. For instance, in the aforementioned case of an annual mileage of 20 000 km (with a petrol car with a fuel consumption 6.0 L/100 km and an emissions rate of 144 gCO_2/km [177]), the avoided fossil fuel use represents a significant step towards GHG mitigation, amounting to approximately 2.88 tCO_2/year.

It is noteworthy that the negative CO_2 emissions potential reported in Table 4 for the DC-SOFC will remain unaffected by any reduction in space heating demand of the average dwelling. Indeed, this negative emissions potential (associated with the fourth case of Table 4) represents an additional contribution to the carbon footprint savings that have already been included with the utilisation of 100% of “green” energy vectors (included in the third case of Table 4). In fact, a transition of the reference space heating system from gas condensing boilers (as in the “base scenario”) to heat pumps (for the thermal demand not covered by the fuel cell micro-CHP) would result even result in an increase in the electrical demand to the DC-SOFC, thereby also increasing its negative CO_2 emissions potential. It thus follows that the results of this study are not confined to the current average (Belgian) dwelling, with an annual gas consumption of 17 000 kWh, as depicted in this work.

Regarding the limitations of this study, it is pertinent to reiterate that the electrical transportation and distribution losses (which can reach approximately 6-7% in the European Union [160]) that are avoided through decentralised electrical production from micro-CHP systems have not been considered in this work. It can thus be surmised that this will serve to

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[1] The French advocacy group négaWatt, which specialises in modelling energy transition scenarios, has identified four types of sufficiency [179]: structural, dimensional, usage and collaborative. Structural sufficiency focuses on organizing spaces and activities to reduce consumption, such as minimizing travel distances. Dimensional sufficiency ensures equipment and facilities are scaled to their actual use, like using appropriately sized vehicles. Usage sufficiency emphasizes efficient use of resources, such as lowering space heating temperature setpoints or extending equipment lifespans, while collaborative sufficiency promotes sharing resources, like carpooling or shared workspaces [180,181].

enhance the decarbonization potential outlined in Table 4 or discussed in this section. However, this effect can be assumed to be compensated by the fact that the negative emission potentials estimated in this work have been calculated without consideration of the entire life cycle of the fuel cells themselves. Indeed, only the negative emissions associated with the utilisation phase of the system have been taken into account (in contrast to, for example, the emissions generated during its construction, transportation, disposal, *etc*). In addition, this work has not considered the detrimental positive GHG emissions associated with the production and transportation of the fuel, in this case biochar (supposedly grown without GHG-emitting fertiliser). Ultimately, these deliberate omissions, which may slightly overestimate the reported negative emissions potential of the fuel cell, are likely to be consistent with the goals of a future low carbon economy in which these very negative emissions fuel cells could play a fundamental role, as demonstrated in this work.

In fact, the main limitations of this study lie in the uninvestigated barriers to the commercialisation of DC-SOFC systems that enable carbon capture at their anode outlet (such as cost or technological readiness) and in the fact that the DC-SOFC considered in this work was assumed to be fully flexible, which has yet to be demonstrated for CHP applications (although this is already partially the case for commercialised SOFCs [166]).

Although decarbonization potentials have not been established systems other than (DC-)SOFCs (because they have been reported in this study as the most promising micro-CHP fuel cell technologies), the methodology used in Table 4 is simple enough to be very easily reproduced for PEMFCs or DFAFCs systems (or for any flexible CHP technology, as long as its LHV electrical and thermal efficiencies are known).

5. Conclusions

From Table 2 it can be concluded that the main advantages of SOFC-based technologies for the decarbonization of the residential heat and power sector lie in their high electrical efficiencies, high quality heat and simple water management. In addition, their high tolerance to impurities, their high fuel flexibility, the possibility of internal reforming that they offer, and their relatively simple composition of the off-anodes gasses, allow the use of carbon-neutral energy vectors, such as biogas (or, more generally, syngas), ammonia, alcohols, which can be considered as promising alternatives because they could be convenient to produce, store and/or transport. Furthermore, this paper has presented in Figure 7 and analysed in Table 4 the decarbonization potential of biomass-fueled DC-SOFCs, which even allow negative CO_2 emissions thanks to the CO_2 purity of the off-anode gasses. Conversely, the slow start-up and low power densities shown in Table 2 are disadvantages of SOFC technologies, which are more specific to mobile applications than to stationary applications (such as CHP). Nevertheless, their cost and durability are still ongoing challenges (as mentioned again in Table 2).

This study has shown that the GHG mitigation benefits of natural gas-fed fuel cell micro-CHPs compared to the overall average individual carbon footprint for an average Belgian dwelling are at best rather limited, even for “ideal” future flexible systems with high efficiencies. In fact, this work has identified carbon footprint savings of only about 0.5 tCO_{2eq}/year for an average dwelling. This should be compared to the current Belgian average individual carbon footprint, which is much higher, i.e. about 15.4 tCO_{2eq}/year. These savings only occur when compared to current marginal electricity emissions (with a rather high emission factor of 456 tCO_{2eq}/kWh_{el}, corresponding to CCGT power plants). Although this marginal emission factor assumption was valid for the whole of 2020 and 2021 in Belgium, as shown by data from Electricity Maps, it is likely to be reduced at some point in the future as the use of natural gas-fired CCGT on the grid is reduced (for the sake of the energy transition). This would further reduce the already small carbon footprint savings potential of natural gas-fed residential fuel cells (or, by extension, other CHPs).

In addition, this work has shown that even “ideal” fuel cell micro-CHPs (powered by fossil fuels) cannot compete environmentally with current grid electricity. Furthermore, it should be considered that this average grid electricity should become even greener every day with the energy transition.

It is therefore essential to implement other GHG mitigation measures. For example, 100% biogas (and/or other climate neutral energy vectors), with the potential help of increased insulation levels, would increase the carbon footprint savings to about 5 tCO_{2eq}/year for the average Belgian dwelling, i.e. 10 times higher than those allowed by the implementation of the natural gas fed “ideal” SOFC micro-CHPs reported in this work.

The electrification of personal vehicles also has significant potential, estimated at 2.88 tCO_{2eq}/year for the selected case study, which is represented by a 20 000 km annual mileage driven by a 6.0 L/100 km petrol car (savings permitted by the reduction of fossil fuel use). Nevertheless, this will still not be sufficient, and (non-technological) behavioural changes are imperative. For instance, as illustrated in the carbon footprint example of Figure 6, dietary habits and savings accounts also have a considerable impact that must be mitigated (representing approximately 2.7 tCO_{2eq}/year per capita GHG emissions).

However, certain fuel cell CHP technologies are capable of facilitating the capture of CO_2 at their anode exhaust. Indeed, pure CO_2 is the sole fuel cell reaction product at the anode of DFAFCs (fed by formic acid, preferably CO_2-neutral and sustainable, as electrofuel and/or derived from biomass). Pure CO_2 is also the sole fuel cell reaction product at the anode of the more efficient DC-SOFCs (fed by solid carbon, preferably biomass). Consequently, negative emissions have been investigated in this work with the example of an efficient future (flexible) DC-SOFC micro-CHP sized based upon the electric demand of the average Belgian dwelling.

Actually, when the DC-SOFC is oversized to also accommodate expected electrification future needs, such as personal vehicle mobility (which will necessitate increased electrical production), it has been demonstrated that its associated negative emissions can easily reach approximately 4 tCO_{2eq}/year for the average Belgian dwelling (including a single electric vehicle with an annual mileage of 20 000 km). This corresponds to slightly more than 1 tCO_{2eq}/year per capita for the considered dwelling (assuming two adult occupants and the current European average fertility rate of 1.6). This exact level of negative emissions per capita in fact represents the minimum required to achieve climate neutrality, as deduced from the unavoidable GHG emissions reported by the IPCC and the projected global population.

Even if the preferred option is to rely on natural sinks rather than unproven technological solutions, exceeding the carbon budget recommended by the IPCC is an unacceptable risk. Consequently, in light of the significant negative emissions potential highlighted in this study, it is strategically imperative for climate policies to consider investments in DC-SOFC technologies, recognising their potentially substantial role in achieving climate neutrality.

Data availability statement

All data that support the findings of this study are included within the article.

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Nomenclature

AFC	Alkaline Fuel Cell
AR6	Sixth Assessment Report (of IPCC)
AwAC	Agence wallonne de l'Air et du Climat (Climate and Air Walloon Agency)
BECCS	Bioenergy Carbon Capture and Storage
CCGT	Combined Cycle Gas Turbine
CCS	Carbon Capture and Storage
CCU	Carbon Capture and Utilisation
DAFC	Direct Alcohol Fuel Cell
DCFC	Direct Carbon Fuel Cell (DC-SOFC, also named SO-DCFC is one type of DCFC)
DC-SOFC	Direct Carbon Solid Oxide Fuel Cell, equivalent to SO-DCFC
DFAFC	Direct Formic Acid Fuel Cell
DME	Dimethyl Ether
EFC	Enzymatic Fuel Cell
GHG	Greenhouse Gasses
HHV	High Heating Value
HT	High Temperature
IPCC	Intergovernmental Panel on Climate Change
IT	Intermediate Temperature
LCA	Life Cycle Assessment
LHV	Low Heating Value
LPG	Liquefied Petroleum Gas
LT	Low Temperature
MC-DCFC	Molten-Carbonate Direct Carbon Fuel Cell
MCFC	Molten-Carbonate Fuel Cell
MEF	Marginal Emission Factor
MFC	Microbial Fuel Cell
MH-DCFC	Molten-Hydroxide Direct Carbon Fuel Cell
(micro-)CHP	(micro) Combined Heat and Power
OEM	Original Equipment Manufacturer
PAFC	Phosphoric Acid Fuel Cell
PEMFC	Proton Exchange Membrane Fuel Cell (or Polymer Electrolyte Membrane Fuel Cell)

SDG	Sustainable Development Goal
SMP	System Marginal Price
SO-DCFC	Solid Oxide Direct Carbon Fuel Cell
SOFC(-O/-H)	Solid Oxide Fuel Cell (with Oxygen-ion/Proton conduction)

Appendix A

A non-exhaustive list of well-known individual carbon footprint calculators has been reported for information and potential comparison purposes here below. All referenced links were last accessed on November 12, 2023, at 16:00 CET.

1. https://calculateurs.awac.be/ - Used in this study.
2. https://nosgestesclimat.fr/
3. https://www.goodplanet.org/fr/calculateurs-carbone/particulier/
4. https://www.rtbf.be/article/mobilite-energie-conso-a-combien-selevent-vos-emissions-de-co2-faites-le-test-avec-notre-calculateur-11074811
5. https://footprint.wwf.org.uk/ (for UK) or https://www.wwf.ch/fr/vie-durable/calculateur-d-empreinte-ecologique (for Switzerland)
6. https://www.footprintcalculator.org/home/en
7. https://coolclimate.berkeley.edu/calculator
8. https://climate.selectra.com/fr/empreinte-carbone/calculer
9. https://neoenea.be/calculateur-simplifie/
10. https://etat.emfro.lu/s3/myimpact
11. https://www.mijnverborgenimpact.nl/en/
12. https://knowsdgs.jrc.ec.europa.eu/cfc (also evaluates impacts against the planetary boundaries and SDGs 3, 6 12, 13, 14 and 15 [178])

Appendix B

The main assumptions reported to the chosen carbon calculator footprint [148] here below. Those case-dependent assumptions have been reported for the reproducibility of the study and to provide examples of the level of detail involved in the chosen carbon footprint calculator.

- Detached house of 200m², 2 adults (children not considered)
- About 2.5 average garbage bags a week, including papers and cardboard, not including organic trash
- Organic trash thrown in an individual compost
- One small petrol car, bought second handed in 2020 and supposably used until 2030
    - 8000 km/year for professional activities
    - 6000 km/year for personal activities
- Distance traveled by plane:
    - 7000 km for professional activities
    - 7000 km for personal vacation
- 14 meals/week (breakfasts not included): 2 red meat-based meals, 3 white meat-based meals, 1 fish-based meal, 5 vegetarian meals, 3 bread-based meat (with cheese)
- 10 drinks/week (that are not tap water)
- Groceries not specifically local but exhibiting bio labels, not frozen, packed (not bulk food), not specifically aligned with the seasons
- The dwelling possesses 2 computers, 2 smartphones (second-handed, changed every 24 months), 2 TVs, 1 fridge, 1 freezer, 1 dishwasher, 1 dryer, 1 washing machine
- 20 hours of High-Definition streaming / week
- 5 pieces of new clothes / year
- 1000 € on a generic savings account

All those assumptions and the carbon footprint calculations are related to the year 2023.

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