

On the Viability of CO₂-Neutral Vehicles Utilizing Carbon-Based Fuels

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0. Abstract

Although battery electric and hydrogen fuel cell vehicles hold great promise for mitigating CO₂ emissions, there are still unaddressed sectors for electrified transport, e.g., the heavy-duty and long-range global shipping industry. Versus conventional combustion fuels, Li-ion batteries and pressurized hydrogen have greater requirements for volume and/or weight per unit energy and require extensive build-out of charging or fueling infrastructure. Here we examine the viability of CO₂-neutral transportation using hydrocarbon or alcohol fuels, in which the CO₂ product is captured on-board the vehicle. Utilization of these widely distributed fuels avoids the immediate need for significant new infrastructure. Solid oxide fuel cells are proposed for energy conversion because they react fuels with pure oxygen, enabling on-board CO₂ capture in a reasonable volume after compression to a moderate pressure of 250 bar; net storage volume is substantially less than of equivalent hydrogen tanks or batteries. During vehicle refueling, captured CO₂ can be off-loaded and subsequently used in fuel production with renewable resources, or sequestered, thereby maintaining carbon-neutrality. Alternatively, such vehicles can be part of a larger CO₂-negative pathway by directly oxidizing biofuels and sequestering the captured CO₂. Storage volume and weight are analyzed for a range of vehicle types, comparing the proposed transportation platform with those of internal combustion, Li-ion battery, and hydrogen fuel cell vehicles; the results suggest that this is the best available option for long-range vehicles.

1. Introduction

Significant inroads have been made in efforts to decarbonize transportation, but commercialization has been mainly limited to light-duty, short-range vehicles, responsible for approximately half of the emissions from the sector.^{1,2} For this subsector, a recent Princeton report, *Net-Zero America* (NZA), which details a range of pathways to reach CO₂ neutrality by 2050, envisions rapid growth in the use of battery electric vehicles (BEVs) alongside major expansion in renewable electricity production.² The majority of the remaining emissions in the sector arise from vehicles considered difficult to decarbonize, e.g., those for freight transport and aviation. Here, sufficient battery capacity is problematic, and the NZA report instead proposes comparatively more scalable hydrogen to be utilized via fuel cell vehicles (H2FCVs).

Where practical, BEVs provide by far the best efficiency utilizing renewable electricity, ~77% delivered to wheels. For the remaining applications, H2FCVs with hydrogen derived from biomass gasification or electrolysis, the main production pathways as slated by the NZA report, are relatively inefficient, ~22% and 33% delivered to wheels, respectively.^{2,3} Such low efficiencies will be problematic because they require the production of much greater amounts of renewable energy upstream. Major hydrogen production and distribution infrastructure must already be in place before such vehicles are serviceable, with carbon-neutral processes eventually needing to take over market share. And while hydrogen offers unparalleled gravimetric specific energy, its

low volumetric energy density is still a challenge, requiring energy-intensive compression to be feasible for most applications.

In this article we assess the viability of CO₂ capture on board vehicles during operation, which can enable the direct use of hydrocarbon and alcohol fuels without CO₂ release. Under this approach, shown schematically in Figure 1, efficiency is considerably improved over H₂FCVs by avoiding the need to first convert such fuels to hydrogen. C-based fuels have the well-known advantage of markedly higher energy density than either compressed H₂ or Li-ion batteries; they also have an existing, well-developed distribution infrastructure. As shown in Figure 1, the CO₂ captured on vehicles could be returned to a CO₂ distribution network where, after sequestration, the cycle is CO₂-neutral for fossil fuels and CO₂-negative for biofuels. Another route, recycling the CO₂ back into fuel via electrolytic processes, is also possible.

The vehicle illustrated in Figure 1 is referred to here as a Carbon Capture Fuel Cell Vehicle (CCFCV). The solid oxide fuel cell (SOFC), shown in the vehicle inlay, is the most desirable choice for vehicle power generation with C-based fuels because it acts as a membrane air separator that combusts fuels with pure oxygen. This is necessary to maintain reasonable tank volumes – combustion with air would dilute the CO₂ with large amounts of nitrogen, and thus require prohibitively large tank volumes, up to 10 times that for CO₂ alone. Although separate fuel and CO₂ tanks could be used, net volume requirements can be further reduced by storing the concentrated CO₂ product stream in the volume made available by spent fuel, e.g., using a tank with a movable partition as illustrated in Figure 1. Note that the use of SOFCs as auxiliary power units, range extenders, or the primary power source in electric vehicles is already seeing rapidly growing interest.^{4, 5}

SOFCs are able to work with minimally-reformed hydrocarbon and alcohol fuels, while providing fuel-to-electricity conversion efficiency of 50–60%;⁶ given a typical electric motor efficiency of 95%, the net fuel-to-wheels efficiency (47–57%) is substantially higher than for typical transportation heat engines (10–40%).² Assuming bioenergy-intensive pathways, the fuel flexibility of the SOFC allows it to operate on higher production efficiency biofuels (up to ~85% for bioethanol)⁷ versus biohydrogen (up to 50%)³ resulting in net renewables-to-wheels efficiencies of ~44% for the CCFCV versus ~22% for H₂FCVs. On the other hand, assuming high electrofuel pathways, the net renewables-to-wheels efficiency of the CCFCV and H₂FCV are similar, but are substantially lower than for BEVs.²

One could argue that a similar carbon impact could be achieved with C-based fuels by simply releasing the CO₂ from internal combustion vehicles (ICEVs) and then using atmospheric CO₂ capture, but this process requires considerable resources – costs may eventually be as low as \$94–\$232 per ton of CO₂, but a more conservative estimate is \$1,000 per ton.⁸ Here, instead of release into the atmosphere, concentrated CO₂ will be captured; as discussed below, on-board compression of CO₂ introduces little energy efficiency or cost penalty.

It will be shown that this approach is viable for a variety of C-based fuels (Section 2), and has significant advantages for longer-range maritime and terrestrial applications for which H₂FCVs are currently under consideration (Section 3). Given existing C-based fuel infrastructure, rapid adoption of such vehicles should be possible, and CO₂ emissions can be continuously decreased as new infrastructure for CO₂ conversion, collection, and/or distribution is built (Section 4).

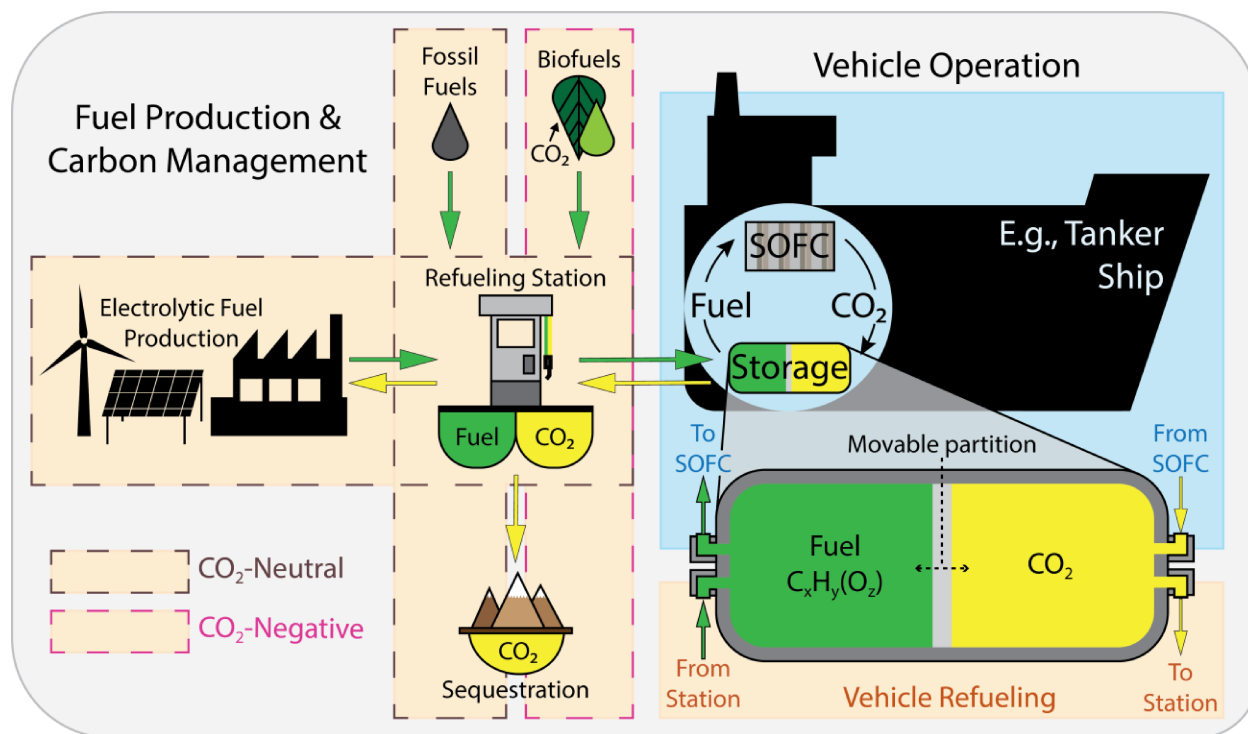


Figure 1. Schematic illustration of a Carbon Capture Fuel Cell Vehicle (CCFCV) and associated infrastructure. The vehicle includes a solid oxide fuel cell (SOFC) for efficient electrical generation from hydrocarbon or alcohol fuels. Fueling is with any of biofuels, fossil fuels, or electrolytic fuels produced using renewable electricity. The captured CO₂ can be stored in a separate tank or in a unified tank with a movable partition, as shown, to minimize net storage volume. After offloading, the CO₂ can either be used in electrolytic fuel production or sequestered. Different infrastructure designs and fuel choices can yield an overall CO₂ impact ranging from mitigatory to net negative.

2. C-based Fuel and CO₂ Storage Requirements

The volume required for the fuel and CO₂ can be assessed based on the known thermophysical properties of the fluids at elevated pressure. First, we consider the properties of compressed CO₂, shown in Figure 2a. Note that the ambient-temperature density increases rapidly with increasing pressure up to 74 bar and reaches a value of 21 mol/L at 250 bar. This pressure, which is commonly used in compressed natural gas vehicles, is assumed in the analyses below, along with a higher value of 700 bar currently being used for hydrogen in H₂FCVs. Also note that 250 bar is routinely reached with standard compressors and that the energy requirement is small, < 2% of the energy released upon producing the CO₂. While higher pressure increases the gas density, it is likely not worth the required increases in pump size, compression energy, and tank reinforcement for most applications.

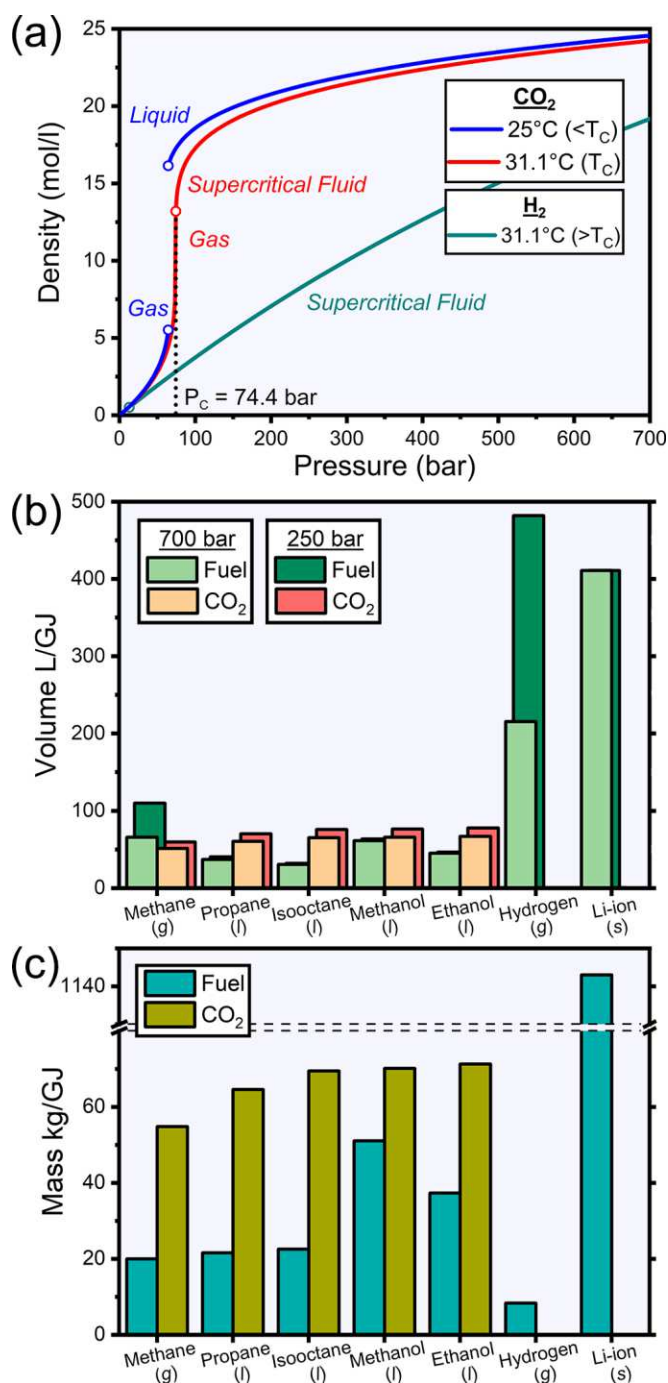


Figure 2. (a) Density of CO₂ versus pressure, at and below its critical temperature (T_c). A similar density is achieved past its critical pressure (P_c). H₂ is also shown for comparison, highlighting the relative ease of compressing CO₂. Comparison of the volumes (b) and masses (c) of different fuels corresponding to 1 GJ* (lower heating value) of stored energy, and of the CO₂ produced by oxidation of the fuels. Values are given for tank pressures of either 250 or 700 bar at 31.1°C. Also shown are values for hydrogen and a representative Li-ion battery. Tabulated data can be found in Tables S1 and S2.

* GJ is used as a convenient measure; note that 1 GJ corresponds to the energy in 31 l of gasoline, approximately the size of a small automobile's fuel tank.

The CCFCV can function on a variety of fuels including hydrocarbons and alcohols. Figure 2b summarizes the requisite storage volumes yielding 1 GJ energy for several candidate fuels, and for the CO₂ produced by combusting the fuel, at both 250 and 700 bar. (Note that the water also produced by combustion would be easily separated from the CO₂ upon cooling the product.) The total volume for a given fuel and pressure can be reduced by utilizing a single adjustable tank versus separate fuel and CO₂ tanks (Table S1), from 30 to 48% depending on fuel and pressure. Considering ethanol, for example, a unified tank reduces storage volume by 38% at 250 bar. Even with separate tanks, a key advantage shown in Figure 2b is that storage volume can be substantially lower compared to hydrogen or batteries.

Figure 2c shows the fuel masses, 20–51 kg/GJ, and product CO₂ masses, 55–71 kg/GJ, with the latter being in some cases more than double that of the fuel. All are shown to be much lower than Li-ion battery mass, 1000–3000 kg/GJ,⁹ but higher than for hydrogen, 8.3 kg/GJ. The increase in mass as fuel is converted is likely not an issue for most terrestrial applications; for example, the mass of a GJ worth of petroleum is ~23 kg and, in the example of a passenger vehicle, a typical GJ-sized tank would increase from ~1% to 2% of total vehicle mass. However, it may be problematic for large aircraft that are designed for low fuel mass upon touch-down.¹⁰

3. Technology comparisons across vehicle classes

To meaningfully assess how CCFCV volume and weight compare with those of other power systems, they must be considered within the context of specific vehicle types. Figure 3 summarizes CCFCV characteristics versus those of ICEVs, H2FCVs and BEVs, using as examples tanker ships and passenger vehicles to represent heavy-duty and light-duty cases, respectively.

Figure 3a shows the power system volume and weight of a tanker ship, a case where the long vehicle range requires a large amount of stored energy. The figure clearly illustrates the challenge for both BEVs and H2FCVs – because of their relatively low energy storage densities, the battery or fuel volume exceeds the available volume, i.e., that which is not dedicated to the cargo. The CCFCV, on the other hand, is clearly a viable path to decarbonized long-range shipping, although the storage volume is ~2x larger versus the incumbent ICEV. SOFC-powered tankers are already under development,⁵ and their conversion to CCFCVs could therefore be more direct than other vehicle classes.

Figure 3b represents the other end of the vehicle spectrum, a light-duty sedan. CCFCV and H2FCV energy systems require reasonably low percentages of total vehicle volume and weight, comparable to ICEVs. Although BEVs have substantially larger requirements, they are still acceptable considering their other advantages, especially their high efficiency when utilizing renewable electricity. All three electrified cases see a substantial fraction of power system volume going to the drive unit (motor, inverter, and differential), and all three contain a battery of differing size for, at minimum, startup and regenerative braking. A key benefit of the CCFCV, its ability to

use high energy density fuel, is not as impactful for this class of vehicle with its lower energy storage requirement.*

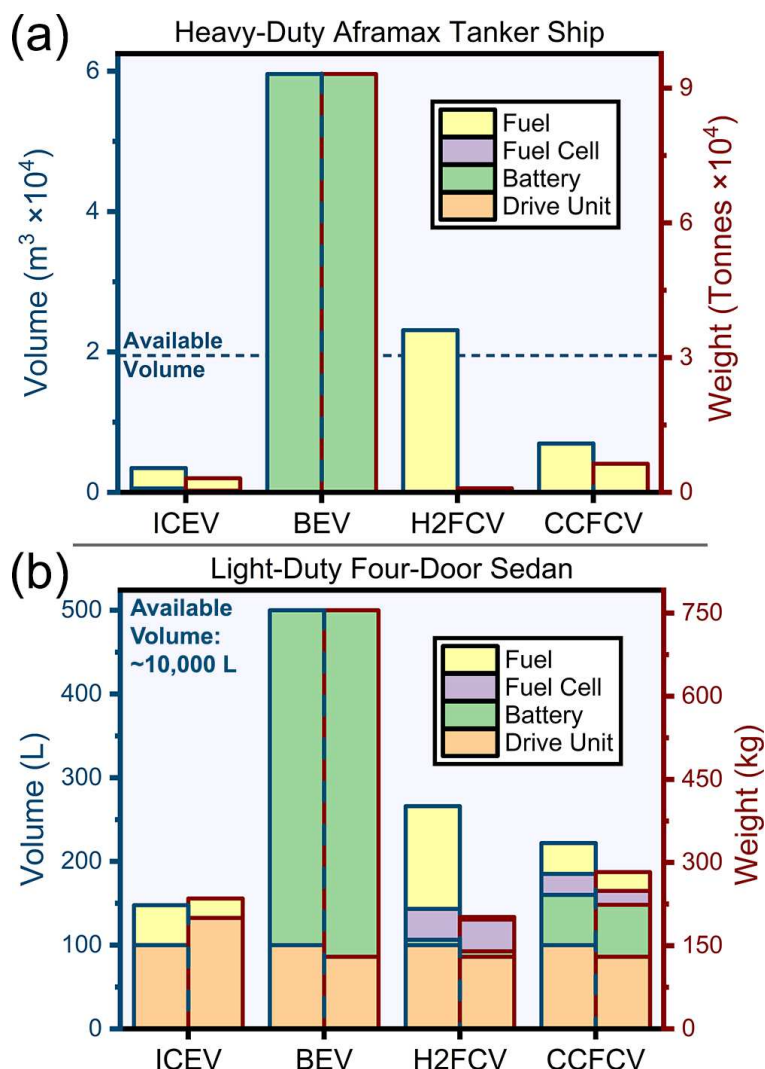


Figure 3. Volume (bars outlined in blue) and weight (bars outlined in red) of the vehicle power system for ICEVs, BEVs, H2FCVs, and CCFCVs with ethanol and 250 bar storage. (a) shows a light-duty sedan of ~500 km range (e.g., Toyota Mirai and Tesla Model S); (b) shows the case of a medium-sized tanker ship (e.g., Wärtsilä WSD42 111K). For (a), the stored energy is by far the predominant portion of total power system weight and volume. Full metrics used for the calculations appear in Tables S3 and S4.

* However, there are other advantages of the CCFCV configuration here, essentially one in which an SOFC serves as a range extender for a BEV.⁴ This design features a substantial reduction in battery pack size compared to full BEVs that could be important for reducing the materials resources required to electrify all light-duty vehicles by 2050.¹¹ The smaller battery makes optimal high-efficiency use of renewable electricity for most travel – short-range trips – whereas for longer-range trips the fuel cell is arguably the highest-efficiency means to utilize C-based fuels.

The above examples broadly represent the array of light- and heavy-duty transportation vehicles; one can expect medium and heavy-duty land vehicles including trains, semitrucks, and busses to have characteristics somewhere between those shown in Figures 3a and 3b. The larger the energy requirement, the more favorable the CCFCV should be compared to BEVs and H2FCVs.

4. Infrastructure, Phased Adoption, and CO₂ Footprint

New infrastructure requirements for adoption of CCFCVs can be much less obtrusive than for BEVs and H2FCVs. The proposed transportation platform can be implemented in stages, as outlined below, with little initial need for new infrastructure before widespread introduction. The engineering challenges will therefore at first be concentrated to the vehicles themselves, e.g., tank design, thermal integration, and SOFC power density (see S2. Technological Challenges). As more vehicles come online, and infrastructure catches up, the overall CO₂ footprint improves from mitigatory, to neutral, and finally to potentially net negative.

A first phase, then, rolls out the CCFCVs prior to any major new infrastructure. Vehicles are fueled where C-based fuel is available, with refueling times akin to those of ICEVs and H2FCVs. Here, the shift from internal combustion engines to SOFCs alone will reduce CO₂ emissions by way of higher fuel efficiency. Early vehicles can still be outfitted for on-board capture, and capture can proceed if the operator expects to refuel at a station capable of CO₂ offloading. However, where this infrastructure is not yet available, the CO₂ can be allowed to bypass the tank; the *catch-22* of needing both infrastructure and vehicles for market penetration, a major detriment to H2FCVs and to a lesser extent BEVs, is thereby avoided. Vehicle cost is of course an important consideration, and so detailed cost analyses are needed; qualitatively speaking, SOFCs are currently more expensive than the proton-exchange membrane fuel cells used in H2FCVs, but should become more comparable as SOFC manufacturing ramps up, especially given that SOFCs do not utilize precious metals.¹² The cost of tanks and compressors should be less for CCFCVs compared to H2FCVs because of the reduced compression requirements for CO₂ compared to hydrogen, but the need for an on-board compressor presents an additional challenge.

The next stage of development would see the implementation of CO₂ sequestration and/or utilization.¹³ Existing infrastructure will still be used to supply fueling stations, but significant expansion of CO₂-related infrastructure and processes will be needed. While this will require considerable investment in plants and pipelines, this is generally true for every net-zero pathway.² Infrastructure cost is a key factor, and while costs have been assessed for CO₂ distribution from fixed sources,¹⁴ realistic cost estimates for the proposed CCFCV platform will require additional analysis. Qualitatively, CO₂ storage and distribution infrastructure should be more cost effective than hydrogen because of the lower compression requirements and elimination of materials embrittlement issues. Note that local distribution could be done relatively inexpensively by retrofitting fuel supply vehicles so that they can return compressed CO₂ to centralized sites.

At all stages, fuel provenance is important, because it helps determine the utilization efficiency and the amount of CO₂ released or captured. Even without CO₂ capture, the carbon footprint can be substantially decreased by shifting from fossil to bio- and electrofuels (and their production can also help alleviate renewable electricity curtailment by being produced during times of grid oversupply). Biofuels may be more desirable, given they can be produced with high efficiency (~85%)⁷ and have the potential to be net negative upon the introduction of any level of

CO₂ sequestration.⁶ On the other hand, carbon-neutral electrofuels, while less efficient to produce (~76%),² have the advantage of locally closing the carbon loop – that is, by equipping fueling stations with electrolysis and fuel production capabilities to utilize CO₂ on-site. If nothing else, fossil fuels could still be conceivably made net neutral were complete sequestration to be implemented.¹³

In summary, vehicles operated on bio-, electro-, or fossil fuels with on-board CO₂ capture could provide a potentially important complement to hydrogen fuel cell and battery electric vehicles, with the practical advantage of system sizes not far from current internal combustion vehicles. The main application will likely be for the decarbonization of heavy-duty, long-range vehicles, for which the high energy storage requirement leads to unmanageably large hydrogen tank size or battery size/mass. Different fueling and CO₂ sequestration/utilization options are available depending on the available local infrastructure, and the requirement for new infrastructure is decoupled from the rollout of vehicles, allowing for optimized pathways towards decarbonization. In its ideal manifestation, an overall carbon-negative cycle can ultimately be achieved by combining renewable biofuels, on-board CO₂ capture, and sequestration. The result is an erasure of emissions from the global carbon ledger, a measure appearing increasingly necessary.

5. Conflicts of interest

The authors declare a competing interest for a patent pending with serial number PCT/US20/44838, regarding the conceptualization of a solid oxide fuel cell in combination with dual-chamber fuel/exhaust storage.

6. Acknowledgements

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