

Carbide-based fuel system for undersea vehicles

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Abstract

In underwater applications such as unmanned undersea vehicle (UUV) propulsion, mass and volume constraints often dictate system energy density and specific energy, which are targeted to exceed 300 Wh L^{-1} and 300 Wh kg^{-1} , respectively, in order to compete with state-of-the-art battery technologies. To address this need, a novel carbide-based fuel system (CFS) intended for use with a solid oxide fuel cell (SOFC) is under development that is capable of achieving these energy metrics as well as sequestering carbon dioxide. The proposed CFS uses calcium carbide and calcium hydride that react with water to generate acetylene and hydrogen as the fuel and calcium hydroxide as a carbon dioxide scrubber. The acetylene is hydrogenated to ethane and then reformed to syngas (carbon monoxide and hydrogen) before being utilized by the SOFC. Carbon dioxide effluent from the SOFC is reacted with the calcium hydroxide to produce a storable solid, calcium carbonate, thus eliminating gas evolution from the UUV. A system configuration is proposed and discussion follows concerning energy storage metrics, operational parameters and preliminary safety analysis.

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Keywords: Calcium carbide; Acetylene; SOFC; UUV propulsion

1. Introduction

Despite wide-ranging efforts to use fuel cells in niche military applications [1–3], the transition is proceeding slowly. This is due in part to the bulky balance of plant (BoP) components and fuel storage requirements associated with fuel cell systems. Mass and volume constraints often dictate system energy density and specific energy, which preferably exceed 300 Wh L^{-1} and 300 Wh kg^{-1} , respectively, in order to compete with battery technologies. In the case of underwater applications, such as unmanned undersea vehicle (UUV) propulsion, the challenges of oxygen storage and maintaining neutral buoyancy of the vehicle are also encountered.

Solid oxide fuel cells (SOFCs), an emerging technology, are being targeted for UUV applications, because of their ability to utilize hydrocarbon fuels without extensive fuel processing. Using logistics-type fuels allows for minimal downtime and rapid refueling between vehicle missions. Proton exchange membrane (PEM) and alkaline fuel cells are also options,

but they require pure hydrogen for reliable performance, and hydrogen storage methods cannot yet compete with liquid hydrocarbon fuels on an energy density basis. While extensive modeling has been conducted on various fuel cell systems for air-independent vehicles [4,5], this paper focuses on a novel carbide-based fuel system [6] for use with SOFCs. A system configuration for a UUV is proposed and discussion follows concerning energy storage metrics, operational parameters and preliminary safety analysis.

2. Methods

As a first step towards investigating this platform as a viable energy section, mass and energy balances were conducted on the process streams for a system operating at steady state. Heat sources in the system are the burner fueled by the SOFC exhaust, the hydrogenation reaction, the carbide and hydride reactions with water, and the waste heat losses from the SOFC stack. The heat sinks are steam generation and steam reforming of methane. There is ample heat available for pre-heating of LOX streams before they enter the fuel cell and burner.

Components masses and volumes were estimated using off-the-shelf values with projections as to how each component

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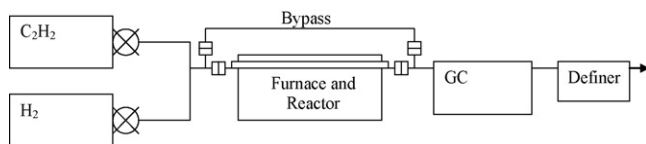


Fig. 1. Diagram of experimental setup for C_2H_2 hydrogenation.

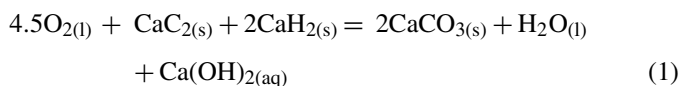
might be optimized for use in UUV. Power production and parasitic losses are estimated from SOFC stack demonstrations and typical balance of plant power demands.

To investigate hydrogenation of acetylene, a microreactor was tested. The reactor was a stainless steel tube of 0.25" inner diameter. One-half grams of a BASF/Englehard catalyst was placed in the center of the reactor and held in place with quartz wool. The catalyst was 1 wt.% Pd on alumina support and had a packing density of 0.82 g mL^{-1} . The flow of reactant gasses, C_2H_2 and H_2 , were controlled by individual rotameters. A definer flow meter (BIOS Definer 220) was used to obtain a reliable measurement of the flow rate of C_2H_2 , and then the desired total reactant flow was obtained by introducing H_2 . A Shimadzu gas chromatograph calibrated for H_2 and C_2H_2 concentrations through a packed column was used to analyze the effluent from the reactor. The use of mass balances allowed the calculation of the concentrations of C_2H_4 and C_2H_6 with the assumption that the effluent of the reactor consists of only the four species H_2 , C_2H_2 , C_2H_4 , and C_2H_6 . The experimental setup is shown in Fig. 1.

3. Theory

3.1. Overall fuel value

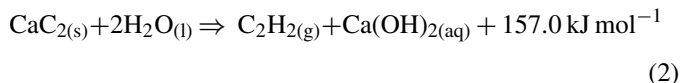
Using a two-to-one molar ratio of CaH_2 to CaC_2 , the overall reaction is



which has an overall heat of reaction of $-3162 \text{ kJ mol}^{-1}(CaC_2)$. On a reactant-only basis, the specific energy is 3000 Wh kg^{-1} and the energy density is 3750 Wh L^{-1} . Assuming an average cell operating voltage of 0.8 V and 80% fuel utilization in the fuel cell, the reactant-only energy metrics are 920 Wh kg^{-1} and 1150 Wh L^{-1} . Overall efficiency would be roughly 30% based on the electrical power generated versus the energy released in Eq. (1).

3.2. Basic reaction chemistry

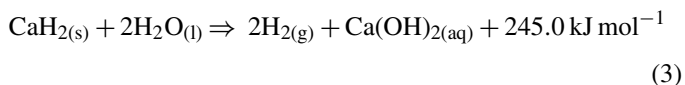
The exothermic reaction of calcium carbide, CaC_2 , with water to produce acetylene gas, C_2H_2 , is given in the following equation:



The energy density of calcium carbide (independent of water) with respect to acetylene production is calculated as

$$\left(\frac{1254 \text{ MJ kmol}^{-1}(C_2H_2)}{64.1 \text{ kg kmol}^{-1}(CaC_2)} \right) (2.22 \text{ kg L}^{-1}(CaC_2)) \times \left(\frac{1 \text{ mol}(C_2H_2)}{1 \text{ mol}(CaC_2)} \right) = 43.4 \text{ MJ L}^{-1}(CaC_2), \text{ LHV.}$$

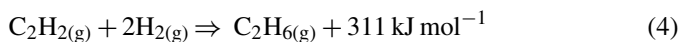
Calcium carbide surpasses the energy density of diesel fuel ($36\text{--}40 \text{ MJ L}^{-1}$) on a lower heating value (LHV) basis, however, it is the acetylene produced by the carbide that is the available fuel. Direct feed of acetylene into traditional SOFC stacks would likely result in carbon deposition, so acetylene must be converted to a suitable feed for the stack [7]. This is accomplished by hydrogenation of the acetylene, followed by steam reforming of the hydrogenation product, ethane, to syngas (hydrogen and carbon monoxide). The conversion requires that both hydrogen and steam be co-generated with the acetylene. Calcium hydride is proposed as the hydrogen source for the hydrogenation step. Its reaction with water is given in the following equation:



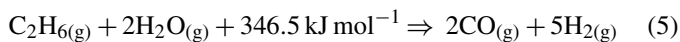
The energy density of calcium hydride, CaH_2 , (independent of water) with respect to hydrogen generation is calculated as

$$\left(\frac{121 \text{ MJ kmol}^{-1}(H_2)}{42.08 \text{ kg kmol}^{-1}(CaH_2)} \right) (1.9 \text{ kg L}^{-1}(CaH_2)) \times \left(\frac{2 \text{ mol}(H_2)}{1 \text{ mol}(CaH_2)} \right) = 10.9 \text{ MJ L}^{-1}(CaH_2), \text{ LHV.}$$

A loss in net stored energy density occurs from using a mixture of CaC_2/CaH_2 versus pure CaC_2 , but the added benefit derived from the following chemistry allows for an entirely closed system, which is advantageous for UUV stealth operation and buoyancy control. A one-to-one molar ratio of CaH_2 to CaC_2 provides perfect stoichiometry for the hydrogenation of acetylene to ethane (Eq. (4)) at $80\text{--}250^\circ\text{C}$ [8–10]. However, excess hydrogen will likely be needed to ensure adequate conversion of acetylene. Therefore a two-to-one molar ratio of CaH_2 to CaC_2 is used:



Steam reforming of ethane to syngas (Eq. (5)) requires heating in a second reactor [11,12] or inside the SOFC at $400\text{--}800^\circ\text{C}$ [13–15]. A small burner can provide the additional heat needed to complete the steam-reforming step as well as pre-heat the fuel for inlet into the SOFC. Exhaust gas from the burner is cooled and returned to the scrubber chamber for CO_2 absorption and water restoration.



The hot reformat (CO and H_2) is consumed in the SOFC with 85% utilization, generating carbon dioxide, steam, electricity,

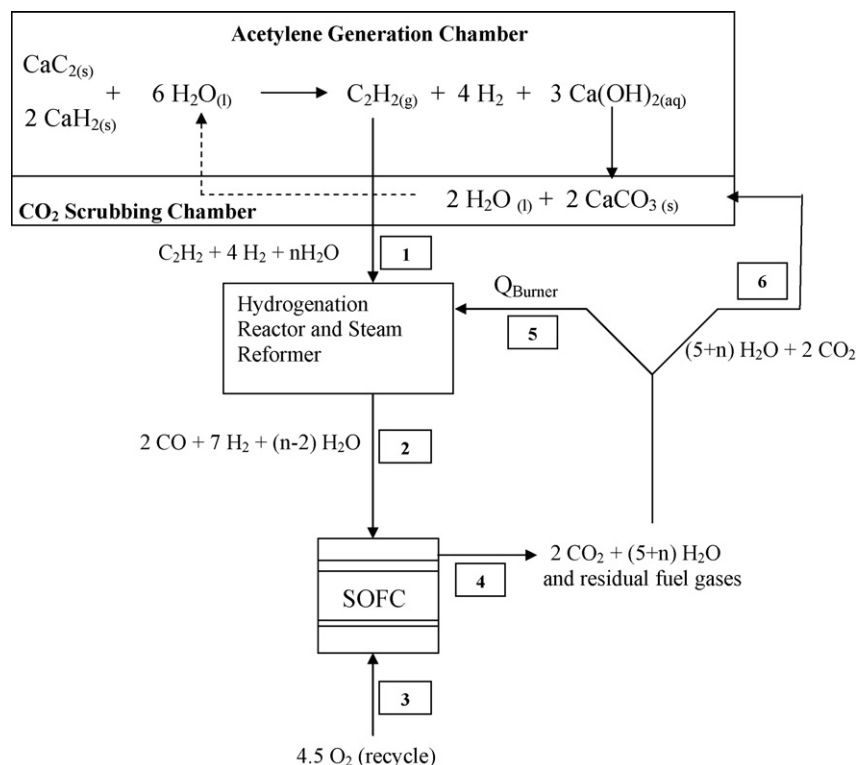
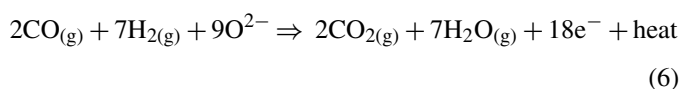
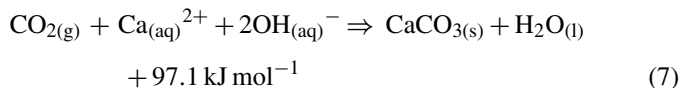


Fig. 2. Carbide fuel system.

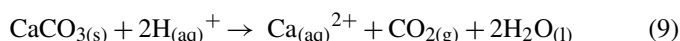
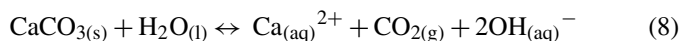
and heat according to the anodic reaction shown in Eq. (6), with a portion directed to the burner.



Product water is stored and reacted with carbide as needed to generate more acetylene and hydrogen. Product CO_2 reacts with the calcium hydroxide, $\text{Ca}(\text{OH})_2$, via Eq. (7).



The water from this reaction is used to generate more acetylene and hydrogen. The calcium carbonate, CaCO_3 , will precipitate out of solution when solubility limits are surpassed. The solubility of CaCO_3 is approximately 0.0015 g/100 cc [16] in cold water (Eq. (8)) with a solubility product $K_{\text{sp}} = 10^{-9}$ and increases with decreasing pH according to Eq. (9).



The reactants CaC_2 and CaH_2 are both basic anhydrides that react with water (Eqs. (2) and (3)) to form a solution containing OH^- ions. The reaction shown in Eq. (7) is favored, ensuring that CaCO_3 will remain as a solid precipitate.

3.3. System design

Fig. 2 is a flow diagram showing the major components and pertinent reactions of the carbide fuel system (CFS). To properly define steady-state operating conditions for the system the following must be considered: thermal management of the acetylene generation chamber (AGC), fuel processing, stream dynamics (gas flows), component sizing and carbon dioxide scrubbing.

There is adequate energy from the exothermic reactions in the acetylene generation chamber to produce steam for the reforming processes, but care must be taken to observe the vapor–liquid equilibrium of water in the chamber. In theory, 6 mol of water are required to generate 1 mol of acetylene and 4 mol of hydrogen. To ensure sufficient steam generation and safe operation, the generation chamber will likely need to operate near 90 °C. In this temperature range, the additional steam produced should help to prevent carbon deposition in the downstream processes. The AGC will require water feed from the carbon dioxide scrubbing chamber to maintain an adequate water level for acetylene generation. If necessary, the water input may also be cooled by heat exchange with seawater to prevent thermal runaway.

3.4. Fuel processing

Hydrogenation reactors are typically designed to perform with dry gas mixtures in the temperature range of 80–250 °C. In the case of C_2H_2 hydrogenation, the effluent stream from the reactor will be heated to induce steam reforming of the ethane as well as raise the temperature of the syngas prior to entering

Table 1

Stream information for the carbide fuel system at steady state with SOFC stack power output of 2.3 kW

Stream	Mole fractions							T (K)
	Flow (mol min ⁻¹)	H ₂	H ₂ O	CO ₂	CO	C ₂ H ₂	O ₂	
1	1.97	.284	0.645	0	0	0.071	0	370
2	2.25	0.500	0.375	0.065	0.060	0	0	973
3	0.513	0	0	0	0	0	1	300
4	2.25	0.093	0.782	0.114	0.011	0	0	1003
5	Heat to reformer from burner = 1480 W, the burner requires 0.117 mol min ⁻¹ oxygen flow							
6	2.325	0	0.858	0.142	0	0	0	400

the fuel cell stack. Also, as stated earlier, an appropriate amount of steam will need to be supplied to prevent carbon deposition. While acetylene decomposition and coking above 400 °C is of concern, high steam levels and the hydrogenation step should help minimize these problems. In addition, by generating steam in the AGC, less heat is required from the recycle stream burner, thus minimizing oxygen consumption and increasing system efficiency and energy storage. Therefore, a helpful initiative would be to develop a hydrogenation reactor that can operate with significant amounts of steam present in the reactant stream. In order to provide proper motive force for the reformat stream, a high-temperature blower may be required. There is ongoing research to develop this technology for related fields that might use SOFC systems with hot recycle streams [17].

4. Results

4.1. Stream dynamics

Table 1 lists estimated stream states required to operate a SOFC stack at a power level of approximately 2300 W (55 A at 42 V) at 80% fuel utilization (of stream 2) and a steam-to-carbon ratio of about 3.0 entering the SOFC. Start-up of the SOFC stack will be accomplished by an external heat source, but once at operating temperature (725 °C), the stack will be self-sustaining and should produce enough internal heat to drive the entire system depicted in Fig. 2.

4.2. Component sizing

Estimated component masses and volumes associated with an SOFC energy system capable of a 24.8 h mission at an average power level of 2.16 kW is given in Table 2. (Components in italics do not factor into totals since they are included in other component specifications.) Several assumptions and scale-up estimates were used to compose Table 2.

Preliminary investigations of the hydrogenation of acetylene to ethane have shown promise. Fig. 3 shows that 90% conversion of acetylene to ethane was seen for furnace temperatures from 100 °C to 160 °C. Most of the leftover acetylene was converted to ethylene with only 1% acetylene in the effluent gas. Gas flow rate into the reactor was kept constant, and as temperature was increased the conversion fell slightly lower. It appeared that the lower residence time overshadowed any benefit from accelerated kinetics at these temperatures. Assuming that this reactor could

Table 2

Component mass and volume estimates

Component	Mass (kg)	Volume (L)
CaC ₂	14	7
CaH ₂	18	10
Generation chamber	10	10
Scrubbing chamber	15	30
<i>Initial water supply (included in chambers)</i>	<i>10</i>	<i>10</i>
Hydrogenation reactor	1	1
Steam reformer	3	3
Burner to heat reformer	2	1
SOFC stack	21	6
Heat exchange lines and piping	10	10
Hot gas recycle pump	4.5	0.6
Pumps for water circulation (4 total)	4	2
Liquid oxygen (LOX)	30	34
LOX tank and vacuum jacket	50	50
Control system	3	1.5
SOFC insulation	9	20
<i>CaCO₃ product (included in reactants)</i>	<i>44</i>	<i>16</i>
<i>Ca(OH)₂ product (included in reactants)</i>	<i>16.2</i>	<i>~8</i>
<i>Net water production (from reactants)</i>	<i>4</i>	<i>4</i>
Totals	195	187

be linearly scaled up from handling 60 mL min⁻¹ to 60 L min⁻¹, then the catalyst mass would be increased from 0.5 g to 0.5 kg. The 0.6 mL reactor would be 0.6 L.

The steam reformer estimates were extended from the work done by Gorte et al. [18]. They determined for ethane reforming a rate near 2×10^{18} molecules_{CO_x} s⁻¹g_{cat}⁻¹ at 500 °C. In this system, up to 0.28 mol min⁻¹ of CO_x species are produced by the

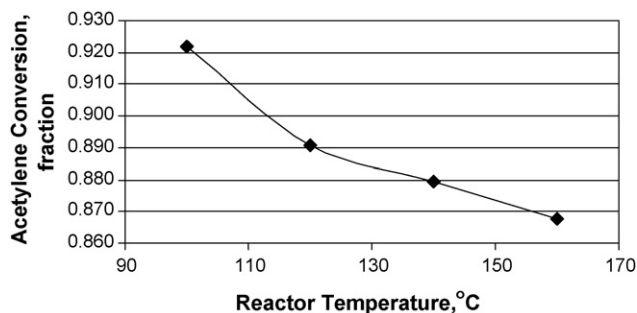


Fig. 3. The conversion of acetylene to ethane as a function of reactor temperature. Gas flow into the reactor consisted of 15 mL min⁻¹ C₂H₂ and 46 mL min⁻¹ H₂. Approximately 1% acetylene was left in the reactor effluent with the rest being converted to ethylene.

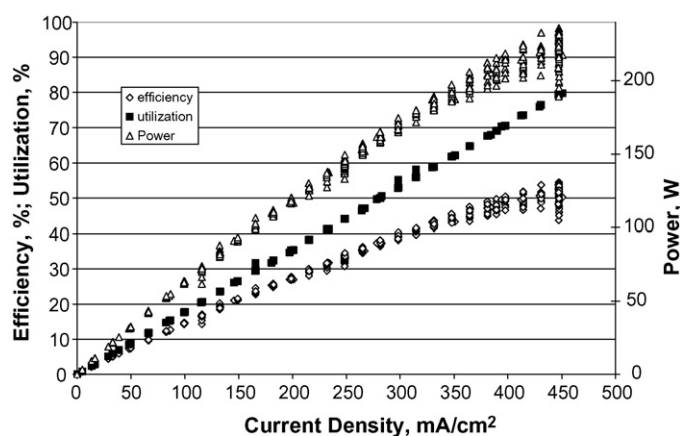


Fig. 4. Performance of a 6-cell Versa Power Systems SOFC stack at NUWCDI-VNPT operating under dodecane reformat and pure oxygen. Efficiency is based upon the lower heating value of dodecane, and the utilization is the fraction of dodecane reformat oxidized in the SOFC stack.

reformer from ethane. The mass of catalyst required would be roughly 1.4 kg assuming linear scale up of this process. To make sure enough space is allocated for the reformer, this is doubled to 3 kg with a volume of 3 L.

For SOFC stack sizing, comparison with a 30-cell Delphi corporation stack was performed because this stack was designed with consideration towards vehicular auxiliary power units. The 30-cell Delphi stack provides 1.6 kW under excess air and hydrogen gas feed streams, and it is 9 kg and 2.5 L [19]. Assuming this 30-cell stack would only achieve 1 kW in the carbide fuel system design at 80% utilization, 2.3 of these 30-cell modules would provide the targeted 2.3 kW. The total stack mass and volume would then be 21 kg and 6 L.

The operational point chosen for the SOFC stack is based upon performance from stacks tested at NUWCDI-VNPT. Depicted in Fig. 4 is a performance plot from a 6-cell SOFC stack from Versa Power Systems. With the furnace temperature at 725 °C and the stack exhaust vented to atmosphere (~1 atm), the stack obtained ~50% efficiency at 80% fuel utilization and 220–230 W. The active cell area was 121 cm², and the feed into the stack was 0.78 mL min⁻¹ dodecane with 2.68 mL min⁻¹ at the anode and 4 L min⁻¹ pure oxygen at the cathode. The steamwater-to-carbon ratio into the steam reformer was 3.6.

Estimates for a high-temperature fan were supplied by R&D Dynamics, who is developing an anode gas recycle fan for coal power plants employing SOFCs [17]. The fan is being designed to operate in a reducing environment at 850 °C with 4–10 in. H₂O pressure rise and efficiency over 40%.

Specifications for the LOX tank are based on a modified design from standard Sierra Lobo tank [20]. Sierra Lobo's standard design uses stainless steel for the entire assembly and individual steel tanks for the LOX dewar and vacuum jacket. The modified design employs an aluminum vacuum jacket, which could also be partially incorporated in the UUV hull structure to minimize mass and volume.

The insulation used to isolate the high-temperature reformer and SOFC stack from the rest of the UUV could be any number of alumina-based materials. Microtherm offers a high perfor-

Table 3
Power distribution

Component	Power (W)
SOFC stack	2300
Water pumps	–40
Hot gas recycle pump	–50
Controls	–50
Total net power	2160

mance insulation, microtherm super A, which has a density of 450 kg m⁻³ and thermal conductivity less than 0.05 W m⁻¹ K⁻¹ [21]. Insulation for the LOX tank is already accounted for in its vacuum jacket metrics.

The expected power distribution for the system is given in Table 3. Based on these estimates, specific energy and energy density are predicted to be:

$$\text{specific energy} = \frac{2160 \text{ W} \times 24.8 \text{ h}}{195 \text{ kg}} = 274 \text{ Wh kg}^{-1},$$

$$\text{energy density} = \frac{2160 \text{ W} \times 24.8 \text{ h}}{187 \text{ L}} = 286 \text{ Wh L}^{-1}$$

While specific energy may not quite be as high as the targeted 300 Wh kg⁻¹, it is still considerably higher than the best performing rechargeable battery, which is the lithium ion battery at 200 Wh kg⁻¹.

At the above power output, estimated heat losses are expected to be in the range of 2.5–3.5 kW. This is at the limit for passive cooling through the hull for the 21 in. diameter UUV. Any significant increase in power may result in excessive heat generation and loss of thermal control inside the UUV. In order to attain higher power levels without overheating, active cooling would be needed or larger UUV platforms could be considered. Active cooling would increase the parasitic load and may also decrease the stealth value of the UUV if seawater is circulated through the vehicle.

5. Discussion

5.1. Direct feed of ethane into SOFC stack

It is well known that SOFCs can benefit from internally reforming methane, but the direct usage of ethane has not been well researched. There is indication that this can be accomplished, but the anode composition may have to be altered to prevent carbon deposition [12–14]. Direct feed of ethane into the SOFC would eliminate the need for the steam reformer, creating more space for fuel and oxidant. If an additional 3 kg of oxygen and 3.2 kg of fuel were added to the system, the energy storage would be increased to:

$$\text{specific energy} = \frac{2160 \text{ W} \times 27 \text{ h}}{198 \text{ kg}} = 294 \text{ Wh kg}^{-1},$$

$$\text{energy density} = \frac{2160 \text{ W} \times 27 \text{ h}}{189 \text{ L}} = 308 \text{ Wh L}^{-1}$$

With SOFC stack technology continually becoming lighter and more power dense, these numbers could be even higher and surpass the 300 Wh kg^{-1} mark. There are also advantages for exploring larger diameter UUV vehicles for this energy system because they can operate at lower power density and offer a greater proportion of available volume for fuel and oxidant storage.

5.2. Carbon dioxide scrubbing

Depending upon the storage methods of the carbide reactants, the CO_2 scrubbing may take place either inside the generation chambers or be physically separated with only the hydroxide solution shared between the chambers. For the former case, the precipitate deposits in the generation chamber. In the latter case, water is pumped from the scrubber chamber into the generation chamber, and warm, Ca(OH)_2 -rich solution is pumped from the generation chamber into the scrubber chamber. The CaCO_3 precipitate deposits in the CO_2 scrubber chamber and is removed at the end of the run. It is not clear at this time, which approach is preferable, and it will likely be influenced by the final carbide delivery method that is chosen.

5.3. Oxygen storage

Liquid oxygen (LOX) storage has yet to be approved for use on board US Navy vessels, but other nations are already using it for fuel cell powered submarines. LOX tanks have yet to be optimized for smaller scale applications such as UUVs. The mass and volume metrics for the LOX tank and vacuum dewar in Table 3 assume that the tank is aluminum and is built into the UUV hull structure. Main concerns involve the required insulation and availability of liquid oxygen in a military setting. Another issue for undersea vehicles is the neutral buoyancy of the vessel. A LOX tank cannot be easily back filled to maintain the same center of gravity on the UUV, so ballasting in other sections of the craft will likely have to compensate for a mass shift from the LOX tank to other product storage areas. Concentrated hydrogen peroxide, perchlorates, and superoxides are all also under consideration as oxygen sources, but each has yet to be optimized for general use in UUVs.

5.4. Safety analysis

A major issue with the use of acetylene mixtures is auto-ignition, the lowest temperature at which a substance will spontaneously combust in a normal atmosphere without an external source of ignition, such as a flame or spark. Most handbooks list acetylene's auto-ignition temperature as 304°C at ambient pressure, but at pressures exceeding 2 atm, it can decompose explosively without any oxidant present [22]. Hence, temperature, pressure, and gas composition parameters must be carefully investigated to determine safe operating regimes in the UUV platform.

The generation of acetylene can be carried out in two ways. Incremental addition of CaC_2 to an excess water supply allows careful control of temperature and pressure in the AGC, mini-

mizing the possibility for auto-ignition. As C_2H_2 is generated, the solution containing Ca(OH)_2 immediately becomes an effective CO_2 scrubber. Alternatively, if water is slowly added to excess carbide as is done in a carbide lamp, CO_2 scrubbing would be severely limited until all the carbide has fully reacted with water. If this latter method is employed, multiple cartridges would need to be used in sequence along with the addition of steam to the effluent gas stream. Finally, CaCO_3 , the end product of the CO_2 sequestration is a non-toxic, insoluble solid that can be used to regenerate the carbide and hydride species. Spent cartridges could be regenerated in a facility capable of producing carbide fuel rods or slurries.

6. Conclusions

Calcium carbide and calcium hydride are an effective means to store chemical energy for use in solid oxide fuel cells. When reacted with water, they produce acetylene, hydrogen and calcium hydroxide. The acetylene produced is catalytically processed in two steps before being introduced into the SOFC as a fuel: (1) hydrogenation to ethane followed by (2) steam reforming to syngas. The hydrogen source for the hydrogenation is calcium hydride in the ratio of two-to-one with calcium carbide. The by-product, calcium hydroxide, serves as a CO_2 scrubber, making for an entirely closed energy system, with only heat transfer through the vehicle hull required.

The energy density and specific energy of the proposed carbide fuel system (taking into account all components) are estimated to be 274 Wh L^{-1} and 286 Wh kg^{-1} , respectively, with the near-term state-of-the-art technology. The proposed oxidant source for the system is liquid oxygen, offers the advantage of fast refueling, but it suffers from bulky cryogenic storage and balance of plant requirements, which add weight and volume to the system. As better oxidant storage methods are developed, the energy storage capacity will likely improve. Advances in SOFC power density and tolerance to ethane will also have favorable impacts on the energy storage capacity of this system design. It is feasible that energy density over 300 Wh kg^{-1} may one day be achievable, however, the safety concerns of the carbide fuel system must be adequately addressed before this design is suitable for vehicular platforms. Auto-ignition and decomposition of acetylene must be mitigated by careful control of the acetylene generation chamber operating parameters (temperature, pressure and gas composition).

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