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Authors	Rusmanis, Davis;Yang, Yan;Long, Aoife;Gray, Nathan;Martins, Kelvin C.;Ó Loideáin, Seán Óg;Lin, Richen;Kang, Xihui;Cusack, Donal Óg;Carton, James G.;Monaghan, Rory;Murphy, Jerry D.;Wall, David M.
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Electrofuels in a circular economy: A systems approach towards net zero

Davis Rusmanis [a,b], Yan Yang [a,b], Aoife Long [a,b], Nathan Gray [a,b], Kelvin C. Martins [c], Seán Óg Ó Loideáin [a,d], Richen Lin [a,b,e], Xihui Kang [a,f], Donal Óg Cusack [g], James G. Carton [a,c], Rory Monaghan [a,d], Jerry D. Murphy [a,b], David M. Wall [a,b,*]

[a] SFI MaREI Centre for Energy, Climate and Marine, Environmental Research Institute, University College Cork, Ireland
[b] Civil, Structural and Environmental Engineering, School of Engineering and Architecture, University College Cork, Ireland
[c] School of Mechanical & Manufacturing Engineering, Dublin City University, Ireland
[d] Ryan Institute, School of Engineering, National University of Ireland Galway, Ireland
[e] Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, School of Energy and Environment, Southeast University, Nanjing 210096, China
[f] Advanced Biofuels and Bioproducts Process Development Unit, Biological Systems and Engineering Division, Lawrence Berkeley National Laboratory, Emeryville, CA 94608, United States
[g] DePuy Ireland Unlimited Company, Loughbeg, Ringaskiddy, Cork, Ireland

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ABSTRACT

Decarbonising the hard-to-abate sectors will be necessary in realising a future net-zero economy. Electrofuels store electricity as low carbon energy vectors such as hydrogen or methane which can be used in areas where electrification is not ideal, and as such can facilitate decarbonisation of sectors such as transport, agriculture, and wastewater treatment. In this study, the production of electrofuels was analysed at an industrial site by storing renewable electricity as green hydrogen produced using electrolysis. The analysis highlighted the need for scale in hydrogen production. The cost of hydrogen was calculated at €8.92/kg when a 122 kW electrolyser operated solely on curtailed electricity generated from the industry site was situated at a 65,000 person equivalent municipal wastewater treatment plant. A subsequent integrated and circular approach to electrofuels production was investigated. The oxygen by-product from electrolysis could be utilised for wastewater aeration and reduce the annual electricity usage at the wastewater treatment plant by 3.6%. Furthermore, the carbon dioxide in biogas generated from sewage sludge could be converted to methane through a Sabatier reaction ($4H_2 + CO_2 \rightarrow CH_4 + 2H_2O$) as a means of carbon capture and utilisation. The hydrogen produced from the 122 kW electrolyser could convert only 40% of the total carbon dioxide (within the biogas) in a biomethanation process, again supporting the argument for larger electrolyser systems with increased hydrogen production. Pyrolysis of digestate to produce biochar was investigated as a negative emissions technology. If pyrolysis is coupled with anaerobic digestion of feedstocks within 10 km of the industry site savings of 42.7 kt CO_2/a could be achieved. In essence, a circular economy approach to electrofuel production could integrate existing electrical, gas and water infrastructure, whilst treating waste, improving the environment, decarbonising agriculture, and storing energy in the form of new low carbon energy vectors for use in heavy transport. Such an approach is vital to progressing future net-zero strategies, however future emissions accountancy processes must adapt to facilitate the benefits of a circular economy.

1. Introduction

Whilst a global consensus was reached through the Paris Agreement to reduce greenhouse gas (GHG) emissions and limit global temperature rise below 1.5 °C, the complex logistics necessary to achieve such decarbonisation cannot be understated [1]. Furthermore, the European Green Deal has subsequently mandated that the EU must be climate neutral by 2050 and that all economic sectors should play a role [2]. However, hard-to-abate sectors such as transport, waste management, and agriculture are often overlooked, despite accounting for a significant proportion of emissions. In Ireland for example, agriculture accounted for 37% of annual GHG emissions in 2021 [3]. This highlights the difficulty in reaching a zero-carbon economy. There is no silver bullet solution to such hard-to-abate sectors. Full electrification will not satisfy all long-distance haulage vehicles nor the production of agri-

* Corresponding author at: SFI MaREI Centre for Energy, Climate and Marine, Environmental Research Institute, University College Cork, Ireland.
E-mail address: david.wall@ucc.ie (D.M. Wall).

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Nomenclature

Abbreviation	Definition
/a	*Per annum*
AD	*Anaerobic Digestion*
AEC	*Alkaline electrolysis cell*
AGI	*Above ground installation*
BOD	*Biological oxygen demand*
CCU	*Carbon capture and utilisation*
CH_4	*Methane*
CO_2	*Carbon dioxide*
Cp	*Capacity factor*
CSO	*Central statistics office*
EU	*European Union*
GHG	*Greenhouse gas*
H_2	*Hydrogen*
HHV	*Higher heating value*
LCOE	*Levelised cost of energy*
LCOH	*Levelised cost of hydrogen*
LHV	*Lower heating value*
NETs	*Negative emission technologies*
O_2	*Oxygen*
PE	*Population equivalent*
PEM	*Proton exchange membrane*
PS	*Primary sludge*
REDII	*Renewable Energy Directive recast*
SMY	*Specific methane yield*
SOEC	*Solid oxide electrolysis cells*
TSS	*Total suspended solids*
TT	*Tube trailer*
VRE	*Variable renewable electricity*
VS/TS	*Volatile solids to total solids ratio*
W	*Watts*
WAS	*Waste activated sludge*
WWTP	*Wastewater treatment plant*

products such as milk and cheese [4]. Holistic approaches are thus required to move towards net zero GHG emissions, facilitated by a circular economy integrating carbon capture and utilisation (CCU) technologies and novel negative emissions technologies (NETs).

1.1. The role of electrofuels in reaching net-zero

One method for progressing towards net-zero can be formed through the production of electrofuels. Recent projections have shown the Irish electricity grid will be operating at 80% renewable electricity by 2030 [5]. Whilst this is crucial, renewable wind and solar power introduces the issue of variable renewable energy (VRE) production which can cause the mismatch of energy production and energy demand. If no means for energy storage is available, electricity is curtailed or constrained [6]. Battery technologies can help alleviate this variability of production but are limited by low storage capacity and low discharge time. Of late, electrofuels (or power-to-X systems) have been mooted as a potential flexible energy storage solution complimenting VRE sources [7]. These electrofuels can be gaseous (hydrogen, methane) or liquid (methanol, Fisher-Tropsch diesel), enabling stable, energy dense, and relatively inexpensive forms of energy storage. Previous studies have shown the viability of hydrogen (H_2) production from VRE, relying on bidding strategies to determine the levelized cost of energy for production of H_2 [8], offering viable pathways for green H_2 in a sustainable green economy.

H_2 generated from the electrolysis of water presents a low carbon energy vector that can accommodate the aforementioned hard-to-abate processes which cannot be fully electrified [9]. H_2 can undergo further upgrading to methane (CH_4) when combined with carbon dioxide (CO_2); this can offset natural gas, maximise the utility of existing infrastructure and increase the share of renewable gas in the gas grid [10]. CH_4 can be further used in the production of fertilisers such as ammonia (NH_3), Fisher-Tropsch diesel or methanol (CH_3OH) [11]. The production of these electrofuels offers a green alternative to fossil hydrocarbon fuels that are widely used, particularly in the transport and agriculture sectors. To ensure a green electrofuel, green renewable electricity must be used in the production. This results in a complex problem requiring a complex solution, with a system-of-systems capable of accommodating variability, capturing carbon, and creating a symbiotic cross-sector relationship. This system-of-systems is not expected to be easy to implement, but this study investigates how society must engage in forward thinking to ensure a sustainable future with sustainable systems.

1.2. Circular approach to electrofuel production

In creating a sustainable economy, a circular approach to energy production can enable the re-use of system by-products and establish a circular resource pathway. This can exemplify a system-of-systems approach, allowing previously independent sectors such as wastewater treatment to be integrated to the wider energy network. Technologies such as electrolysis produce H_2 and oxygen (O_2), with the O_2 fraction often vented due to its market value not justifying the costs of storage and transport [12]. The capture and utilisation of this O_2 by-product has scarcely been investigated in literature. The investigation of electrofuel production and the utilisation of potential by-products (such as O_2), or the sizing of such systems in real-world applications has not been previously described in detail. Previous literature studies from Gray et al. [19], Lung et al. [20], and Ruggles et al. [21], have focused primarily on the function of electrofuels in the wider energy network.

It has been shown that in conventional wastewater treatment plants (WWTP), the aerobic treatment processes typically contribute up to 50% of the total energy demand of the WWTP [13]. Rusmanis *et al.* demonstrated how the by-product O_2 could replace air used to aerate the activated sludge process [14]. This would reduce the overall energy demand of the facility by a factor of 5, leading to substantial cost savings and GHG reductions [15]. Potential commercial viability for co-location of H_2 and O_2 production at the WWTP has also been previously assessed, with evidence of economic benefit for both electrolysers and the WWTP over a 25-year assessment period [18]. Furthermore, if the water is sufficiently treated it can be supplied back to the electrolyser, again supporting the circular economy approach. During the wastewater treatment process, nutrient rich biomass (primary sludge (PS) and waste activated sludge (WAS)) is removed from the contaminated waters and is often disposed of in landfills, or burned, releasing fugitive emissions [16]. Some WWTPs are equipped with anaerobic digestion (AD) systems, using a consortium of microbes to degrade the sludge and generate biogas. This biogas can be used in a combined heat and power unit to generate heat and electricity that can again be used onsite to offset the use of fossil fuels. Further opportunities for carbon capture and utilisation (CCU) can also be unlocked at the WWTP. Technology coupling such as AD and biomethanation (biological methanation combining H_2 and CO_2) can provide for carbon sequestration. Pilot scale demonstrations such as the "Electrochaea BioCat" plant have already demonstrated the potential symbiosis between a power-to-gas facility and a WWTP [17].

Wu et al. [22] and Lin et al. [23] previously assessed the mass and energy balance and GHG emissions, respectively, for a circular

cascading system that integrated anaerobic digestion (AD) and biological methanation to upgrade biogas to high purity CH_4. These studies also focused on pyrolysing the digestate from AD to produce syngas, pyro-oil and pyrochar and concluded that significant benefits could be attained in using biochar as an additive to the biomethanation process, promoting further symbiosis across the systems. Carbon neutrality will require negative emission technologies (NETs) to capture and store surplus carbon in the atmosphere [24]. NETs such as pyrolysis of the digestate from AD to produce biochar can have multiple functions, serving as a soil enhancer for increased crop productivity and a long-term carbon sequestration pathway.

1.3. Aims and objectives

In this study, the authors propose a novel cascading, circular economy system, that is based on the production of electrofuels through the capture of otherwise curtailed electricity. The aim of the system is to provide opportunities for improved decarbonisation of the hard-to-abate sectors, such as transport, agriculture, water, and the energy sector. The system-of-systems approach, as illustrated in Fig. 1, is proposed to demonstrate how the hard-to-abate sectors could be tackled in a future strategy that strives to reach net-zero by reducing multi-sectoral emissions.

The analysis herein considers a feasibility study of the proposed system at a local industrial site in Co. Cork, Ireland, where two existing 3-MW_e wind turbines are located onsite, in close proximity to a WWTP of approx. 65,000 population equivalent (PE), with adequate gas and electricity infrastructure situated in the locality. The proposed system underlines the importance of situating electrofuel systems at opportune locations such as municipal wastewater treatment plants, which are frequently co-located with AD plants to process sludge waste from the WWTP aeration process. Municipal WWTPs exist in almost all urban locations and are often situated near existing grid infrastructure (gas and electricity), making WWTPs strategically optimal locations to produce electrofuels, synergising the three sectors of water, gas and electricity [25].

The potential end use of the H_2 and CH_4 electrofuels focuses on decarbonising the local transport network, in the form of hydrogen fuelled buses or a potential hydrogen ferry operating in the local harbour. Upgrading the carbon dioxide (CO_2) fraction of biogas from the AD process to biomethane can significantly increase the yield and simultaneously incorporate CCU technology into the circular economy system. To further expand the circular economy system the model also incorporates NETs through the integration of the pyrolysis of AD produced digestate. Additional feedstock that is available in proximity to the industrial site is also be accounted for to further reduce the potential for GHG emissions of the locality.

To advance beyond the state of the art in the development of the electrofuels in a circular economy concept, the following objectives are proposed for this study:

- Examine the suitability of storing renewable electricity as green hydrogen in terms of application (end-use) and cost (€/kg H_2) at the proposed industry site;
- Explore opportunities to decarbonise the wastewater treatment process by integrating an electrolyser and recycling oxygen for aeration;
- Determine the potential for carbon capture and utilisation (CCU) from the coupling of anaerobic digestion and biomethanation technologies in the proposed system;
- Investigate the potential for negative emission pathways through the pyrolysis of digestate;

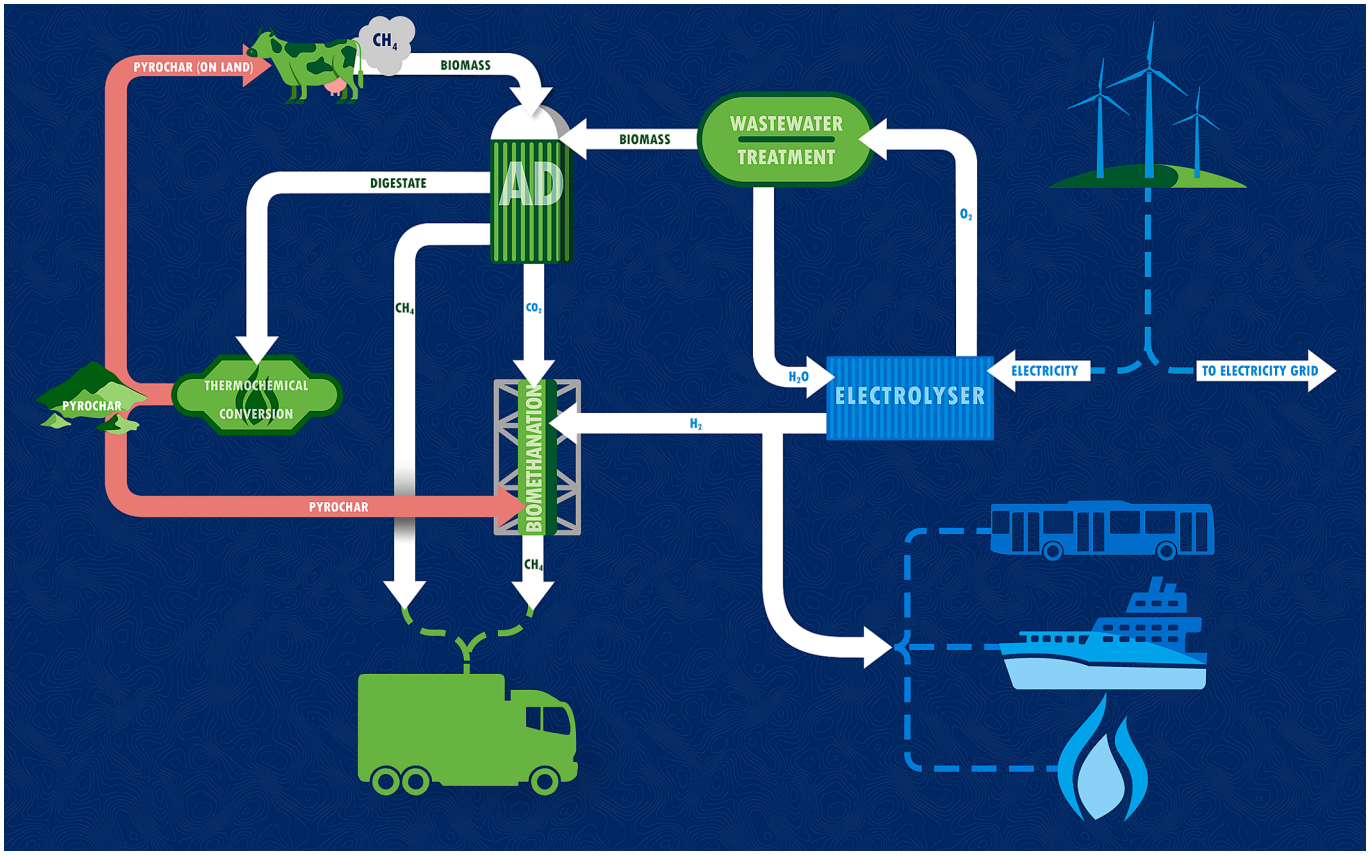


Fig. 1. The proposed circular electrofuels production system.

- Discuss the sustainability considerations to be addressed when implementing a circular economy approach to electrofuel production.

2. Methods

The following section presents the methods used in assessing the electrofuels production within the proposed circular system (as per Fig. 1). The section follows the flow of energy through the proposed system. The investigation starts at the industrial site capturing the surplus wind resource. The assessment of the available wind resource subsequently determines the electrolyser size. In turn, the hydrogen and oxygen resource potential are determined by the chosen electrolyser. This will allow for the evaluation of hydrogen as a potential transport fuel (for buses and a ferry), as well as the potential for emissions reduction in the wastewater treatment process through valorisation of the oxygen. Finally, the wastewater sludge resource is calculated for anaerobic digestion, the biomethanation potential (combination of H_2 and CO_2) is analysed and the subsequent negative emission technology (pyrolysis of digestate) is also investigated.

2.1. Potential to store surplus variable renewable electricity as green hydrogen

To assess the ability to generate green hydrogen from surplus electricity, an industry site with two existing 3-MW_e wind turbines located in the Lower Harbour in Co. Cork, Ireland was selected as a case study for analysis. Granular data was obtained for the wind turbines, for the year 2019, which included for daily generated electricity, electricity demand on-site, and daily exported electricity to the grid. The data implied that the total electricity generated was 15.3 GWh_e/a with an average capacity factor (the ratio of actual output to the maximum possible output) of 28.7%. Of this total, ca. 12.8% (1.96 GWh_e/a) could be considered surplus to onsite requirement. The majority of the electricity generated was used by the industry across various onsite electrical demands.

2.2. Design of electrolyser for the industrial site

The electrolyser is the core component of an electrofuel system. The capital cost of the electrolyser and the cost of electricity are the main factors affecting the overall fuel production cost [26]. Electrolyser costs have reduced by more than 60% in the last decade and are anticipated to decline by more than 50% after 2030 [27]. Currently, the three most common electrolysers for green hydrogen production are the solid oxide electrolysis cells (SOEC), the alkaline electrolysis cells (AEC), and the proton exchange membranes (PEM). Of the 153 completed or planned power-to-gas (electrofuel) projects that are reported in literature, half of the projects have included for PEM electrolysers whilst the other half use AEC systems [10]. An overview of electrolyser characteristics, operating temperature, operating pressure, current density, efficiency, and start-up times are listed in Table 1.

In selecting an electrolyser for the proposed industry site and the existing surplus electricity resource, a PEM system was considered optimal due to its ability to operate at high electrical current densities and variable power supply rates. The two turbines can theoretically produce up to 35,200 kg H_2 annually by utilising all the surplus electricity (see Box S1 in supplementary data). However, due to the intermittent nature of VRE, capturing all of it using an electrolyser would not be viable; a system that would be big enough to capture all the excess energy of the winter months would be lying idle for long periods in the summer, producing hydrogen at very high prices. Thus, choosing the appropriately sized electrolyser requires consideration in terms of how much excess energy could be captured and finding the optimal point between electrolyser capacity, operating hours, and cost. Using the hydrogen production model developed by Gunawan et al. [30] and the analysed hourly wind data from wind turbines, the ideal electrolyser size for the industrial site could then be assessed.

Table 1
Summary of Electrolyser Technologies in 2020.[a]

	AEC	PEM	SOEC
Cell component	Electrodes, membrane diaphragm, alkaline solution	Electrodes, solid polymer electrolyte	Electrodes, ceramic electrolyte
Charge carrier	OH^-	H_3O^+/H^+	O^{2-}
Electrolyte	NaOH or KOH Alkaline solution	Solid Polymer	Ceramic
Operation parameters			
Operating Cell Temperature (°C)	60–90	50–80	700–900
Operating Pressure (bar)	10–30	20–50	1–15
Current Density (A/cm^2)	0.25–0.45	1.0–2.0	0.3–1.0
Flexibility			
Cold Start-up Time	1–2 h	5–10 min	Multiple hours
Load Range (% of nominal load)	20–100	0–100	−100/100
Warm Start-up Time	1–5 mins	< 10 secs	Up to 15 min
Efficiency			
Process efficiency (% HHV)	65–82	67–82	80–90
Durability			
Cell Stack Lifetime (k hours)	55–120	60–100	< 10
Efficiency degradation (%/a)	0.25–1.5	0.5–2.5	3–50
Economic			
Investment cost (€/kW)	800–1500	1400–2100	>2000
Maintenance cost (% of investment cost/a)	2–3	3–5	NA
Stage of technology	Mature	Commercial	Demonstration
Comments	• Long cold/warm start-up time can be a major disadvantage if the electrical input is renewable curtailed electricity	• Act dynamically in managing VRE sources Heavy reliance on the availability of rare metals (e.g., iridium, platinum)/ expensive materials. The degradation of the catalysts leads to a short lifetime	• High energy efficiency Low emission rate

[a] *: Adapted from [17,28,29]; HHV: higher heating value.*

2.3. Potential end uses for the green hydrogen

The green hydrogen produced could potentially be used directly as a transport fuel, or, injected directly to the existing natural gas grid which is accessible in proximity to the industrial site. At present, only small blends of hydrogen are permitted to the grid and many EU countries have stated a limiting range of 0–10 vol% [31]. It is expected that more injection of hydrogen to the existing natural gas grid will become

prevalent in the next decade. For instance, Ireland has a target to introduce 13 vol% of hydrogen to gas network by 2050 [32]. The closest above ground installation (AGI) injection point to the industry site, at which the hydrogen could be injected to the existing gas grid, was located at a 6-km distance away and adjacent to the local WWTP. Preliminary modelling determined that it would be technically feasible to inject hydrogen to the local gas grid as volumes would be below current blending limits in Ireland (<0.1 vol H_2%). However, injection to the grid was calculated to incur an additional cost to hydrogen production assessed of €2.25/kg H_2 for transportation of the compressed gas from the industry site to the location of the AGI; this was assessed using Equations 1–4 below and the assumptions as described in Gunawan et al. [30].

$$LCOH_{trans} = \frac{\sum_{t=0}^{t=\tau WHS} \frac{C_{Inv,TT}}{(1+r)^t} + \sum_{t=0}^{t=\tau WHS} \frac{C_{OM,TT}}{(1+r)^t}}{\sum_{t=0}^{t=\tau WHS} \frac{M_{H_2}}{(1+r)^t}} \quad (1)$$

where;

$LCOH_{trans}$ is the levelised cost of transported hydrogen; $C_{Inv,TT}$ is the sum discounted current value of investment in the tube trailers (TT); r is the investment return; t is the number of years; and $C_{OM,TT}$ is the operation and upkeep expenses over the system lifespan per kg of H_2.

$$C_{OM,TT} = \left(\left(C_{O,TT} + C_{M,TT}\right) \times L_{AGI} \times N_{trip} \times 2\right) + C_{R,TT} \quad (2)$$

Equation (2) evaluates the complete operation and upkeep costs for the tube trailers. In the equation; $C_{O,TT}$ are the operational costs per kilometre for the tube trailer, $C_{M,TT}$ are the tube trailer upkeep costs per kilometre, L_{AGI} is the minimal distance between the wind farm and a grid injection location, N_{trip} is the sum trips carried out per annum(the return travel is represented by 2) and $C_{R,TT}$ is a ten-year tube trailer re-examination costs.

$$N_{trip} = \frac{365}{\frac{t_{SV}}{24}} \quad (3)$$

$$t_{SV} = M_{TT}/m_{H_2} \quad (4)$$

In order to reduce travel costs for each wind farm, N_{trip} denotes the mean storage period (t_{SV}), based on the mean hourly production in kg/hr (m_{H_2}), and the size of the tube trailer (M_{TT}), as shown in Equation (3) and Equation (4).

It is also possible to use the hydrogen generated to produce other synthetic gaseous energy carriers such as methane, or liquid energy carriers such as methanol (CH_3OH) [33]. This approach will be investigated in Section 2.5 as a means of CCU, that is, combining hydrogen with carbon dioxide to produce methane or methanol.

2.4. Oxygen recovery in the wastewater aeration process

The circular economy model can be realised through the integration of the electrolyser at a municipal WWTP which is typically a large energy consumer in municipalities. Oxygen from electrolysis is a valuable resource for the energy-intensive aerobic treatment processes commonly adopted at WWTPs. The aeration energy demand of conventional WWTPs using activated sludge systems, accounts for ca. 50% of the total plant energy due to the pumping of large volumes of compressed air into the aeration tank [13]. When it is considered that oxygen comprises only 21% (volumetric ratio) of air, then pumping air equates to pumping 79% of redundant gas volume (predominately nitrogen) which is not required for aeration. As such, pumping can be reduced by a factor of 5 if pure oxygen is used in aeration. Oxygen from electrolysis can be used at the WWTP to reduce energy costs (pumping O_2 in lieu of air) and as such the carbon footprint associated with the aeration processes [34]. This approach adds value to the oxygen by-product, which may otherwise be vented-off due to lack of cost-effective local solutions. Situating the electrolyser at the WWTP creates an opportunity to partially subsidise hydrogen production and increase its commercial viability [18].

In practice, the oxygen levels in the aeration tank of the WWTP must be maintained constantly to sustain biological activity. Equation (5) presents a standard design formula adapted to evaluate the sum oxygen demand for an activated sludge system [35].

$$OD = 0.75{*}Q{*}(BOD_i - BOD_e) + 2{*}V_A{*}\frac{MLSS}{1000} + 4.3{*}Q{*}$$

$$(Amm_i - Amm_e) - 2.83{*}Q{*}[(Amm_i - Amm_e) - N_e] \quad (5)$$

In the equation, OD is the WWTPs hourly oxygen demand, (g/h); Q is the flowrate at inlet of the WWTP, (m^3/h); V_A is the volume of the aeration tank, (m^3); MLSS is the mixed liquor suspended solids in the aeration tank (g/L); BOD_i and BOD_e are the inlet and outlet concentrations of BOD (g/m^3); Amm_i and Amm_e are the inlet and outlet concentrations of ammoniacal nitrogen (g/m^3), and N_e is the exit flowrate of nitrate nitrogen (g/m^3).

When raw unsettled sewage is processed with extended aeration, the factor of '0.75' changes to '1.0' in Equation (5). This is due to omission of the primary settling from the treatment process. These calculations ignore the efficiencies of the shaft and the motor, among other losses. As such, the equations are a simplified estimate of the reality. For higher resolution results, a revised assessment of each individual WWTP is necessary for exact oxygen demand and sludge production is necessary.

2.5. Carbon capture and utilisation through biological methanation

Anaerobic digestion at WWTPs is commonly used for the treatment of sewage sludge. This sludge primarily comprises of primary sludge (PS) from the primary sedimentation tank (if one is employed) and waste activated sludge (WAS) from the aeration basin. The primary sedimentation tank removes approximately 50% of the solids and produces a relatively rich primary sludge which offers relatively high gas yields in the AD process (Equation (6).

Equation (6): Primary sludge removal

$$PS_{removed} = \frac{50\% \times Q \times TSS \times 24}{1000} \quad (6)$$

In the equation, PS is the removed primary sludge (kg DS/d); Q is the municipal WWTP inlet flowrate (m^3/h); TSS is the municipal WWTP inlet total suspended solids (g/m^3); 50% is the TSS rate of removal from the primary tank. The second biomass removal occurs during the extended aeration process, where WAS is generated as a by converting organic material into cell biomass (Equation (7).

Equation (7): Waste activated sludge production

$$WAS_{produced} = 0.75 \times \frac{Q \times (BOD_i - BOD_e) \times 24}{1000} \times 0.6 \quad (7)$$

In the equation, WAS refers to the produced waste activated sludge (kg DS/d), 0.75 denotes the 25% of the BOD removed from the wastewater due to the associated 50% TSS removal during primary sedimentation; Q is the municipal WWTP inlet flowrate (m^3/h); BOD_i and BOD_e are the inlet and outlet concentrations of BOD (mg/L), and 0.6 refers to the waste sludge output in kgDS/kg $BOD_{removed}$. The properties of these can be found in Table 2.

To optimise the positioning of the electrolyser at the wastewater treatment facility and to complement oxygen utilisation in the waste-

Table 2
AD process yields for primary, and waste activated sludge.

Primary Sludge			Waste Activated Sludge		
SMY	345	L CH_4/kg VS	SMY	87	L CH_4/kg VS
CO_2	33	%	CO_2	50	%
CH_4	67	%	CH_4	50	%
VS/DS	77	%	VS/DS	48	%
Ref.:	[36,37]		Ref.:	[38]	

SMY – Specific Methane Yield.

water treatment process, the green hydrogen generated can also be used onsite to upgrade biogas from the AD process to methane via the biological hydrogen methanation process utilising the Sabatier reaction (Equation (8).

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O, \Delta G_0 = -165kJ/mol \quad (8)$$

The reaction is facilitated by hydrogenotrophic methanogenic archaea [31]. As per the equation, a source of CO_2 is required. Biogas produced by sludge digesters at municipal WWTPs can serve as a biogenic source of CO_2.

2.6. Integration of pyrolysis as a negative emissions technology

Negative emission technologies (NETs) will be necessary in meeting future climate mitigation scenarios [39]. Thermochemical treatment of digestate in the form of pyrolysis under an oxygen free environment can offer a potential NET pathway. Digestate comprises of water, microbial biomass and slow to degrade substrates, which can be dewatered, and applied to agricultural land as a biofertiliser [40]. Pyrolysis of digestate results in the production of biochar (a stable carbon-rich charcoal like material). Biochar can be applied as a soil enhancer for land-based crop productivity, increasing soil organic content, leading to increased photosynthesis and increased draw-down of CO_2 from the atmosphere, establishing long-term storage of carbon [39]. Additionally, pyrolysing digestate can reduce the agricultural land requirement for digestate application, while producing additional energy in the form of syngas and pyro-oil [41]. A NET system incorporating pyrolysis can enable the production of biochar as a means of long-term CO_2 sequestration from the atmosphere (Fig. 1). The addition of the process to the existing circular electrofuel production system would maximise the potential for GHG emission reduction.

The key to realising this NET lies in the technology for the pyrolysis of organic feedstock. The stability of biochar depends on the elemental oxygen-to-carbon (O/C) ratio. Biochar requires an O/C ratio below 0.2 for it to possess an approximate half-life of over 1000 years. However, biochar with O/C ratio of 0.2–0.6 has a half-life of 100–1000 years [42]. Biochar can also be used as a carbonaceous additive, which can be recycled back into the digester and into the biological methanation process to enhance CH_4 production through potentially improved direct microbial electron transfer efficiency [43,44]. Literature has shown that biochar (derived from woody biomass) added to the digestion of seaweed enhanced the CH_4 yields by 17% [41]. Furthermore, the produced syngas from the pyrolysis process could be a supplementary substrate for biological methanation, due to high H_2 and CO_2 content [45]. Implementation of this NET would synergise with the majority of the system components, simultaneously boosting biomethane yields, reducing digestate amounts and increasing net energy outputs. Wu et al. proposed a bespoke circular cascading system with AD as the focal technology, utilising electrofuels and biochar, demonstrating a reduction of 11% in the digestate amount and a 70% increase in biomethane production [22]. The study also claims that such an integrated system could achieve a 70% increase in the net energy output (in the form of methane), as compared to traditional anaerobic digesters with amine scrubber biogas upgrading technology.

2.7. Emissions reduction by integrating anaerobic digestion with pyrolysis

The viability of the NET for GHG emissions reduction depends on the availability of feedstock in the surrounding area for the AD process. This determines the quantity of digestate for the thermochemical process (pyrolysis producing biochar). Increasing the quantity and quality of the pyrolysis products can be achieved through co-pyrolysis of the WWTP sludge and other organic feedstocks available in the locality.

In an Irish context, typical feedstocks for AD include cattle or pig slurry, grass silage, and food waste [46,47]. The mixing of different AD feedstocks and process conditions for pyrolysis will both impact the biochar yield and its carbon content. For example, pyrolysis of sewage sludge is stated to have a biochar yield of 40.2–54.5% of the feedstock dry weight and an average carbon content of 18.1–31.5% [48,49]. In contrast, pyrolysing agricultural residues was shown to offer a biochar yield of 23.7% of the feedstock dry weight and an average carbon content of 55–70% [50].

A simple assessment of GHG emission savings through biochar production is presented herein. A reasonable assumption is that based on 100 t dry sewage sludge, at least 26.7 t CO_2 (=100 t dry solids in sewage sludge × 40.2% biochar yield × 18.1% carbon content in feedstock × 3.67 tCO_2/t C) can be sequestered through biochar production via the combined AD and pyrolysis system. Furthermore, the biochar can improve soil fertility through increased soil organic content, enhancing crop growth and yield with the increased photosynthesis efficiency, leading to additional CO_2 sequestration from the atmosphere [51].

3. Results and discussion

Based on the methods proposed in Section 2, the potential for electrofuel production in a circular economy is now assessed. The following results and discussion section presents the calculations for the industrial site in terms of hydrogen and oxygen production, electrolyser specification, the end-uses for both hydrogen and oxygen, the potential for CCU and the emissions savings from the use of NETs.

3.1. Wind resource potential from the industrial site

The percentage of total wind energy dispatched down as compared to the total produced electricity in Ireland in 2019 was 11.4% (ca. 1,448 GWh out of 12,656 GWh) [6], which is not dissimilar to the ratio of surplus electricity (12.8%) at the selected industrial site. This excess electricity resource at the industrial site could now be considered for green hydrogen production through water electrolysis (Fig. 2). Two end uses were proposed for the hydrogen: 1) direct injection to the existing gas network and 2) utilisation as an advanced transport fuel. A preliminary analysis indicated that the maximum resource of hydrogen that could be generated at the industrial site was 392,000 m^3 H_2/a or 35,200 kg H_2/a (Box S1 in supplementary data).

3.2. Electrolyser specifications for the industrial site

Utilising the model developed by Gunawan et al. [30], the optimal electrolyser size was calculated to be 122 kW as per Fig. 3. Such an electrolyser would have a capacity factor of 74% and produce up to 17,100 kg H_2/a (ca. 190,300 m^3/a) at 60% (LHV) efficiency, representing 48.5% of the aforementioned theoretical resource of hydrogen that could be produced at the site (Box S1 in supplementary data). The Levelised Cost of Hydrogen (LCOH) in this case, taking account of the electrolyser size and resource of surplus electricity, was calculated at ca. €8.92/kg H_2, as seen in Fig. 3.

The 122 kW electrolyser proposed for the industry site would produce hydrogen at the lowest possible cost of a range of scenarios tested. However, the volume of hydrogen produced is relatively small and the cost is high; €8.92/kg H2 is equivalent to €0.27/kWh, well above commercial natural gas prices at industry scale (ca. €0.03/kWh in 2021) though not dissimilar to prices in 2022 as a result of the geopolitical events in Ukraine. An alternative strategy could be carried out to adopt a larger system and increase the output, although that would result in an even higher LCOH (as per Fig. 3). This is further illustrated in Fig. 4, which displays the excess electricity in red throughout the year from the two turbines at the industrial site. The electricity resource captured by the 122 kW electrolyser is shown in green. A decision could be made to increase the quantity of electricity captured by choosing a larger, more expensive electrolyser. Furthermore, an electrolyser at 122 kW is not a commercially available option, the closest available being a 140 kW system which could be obtained by a commercial manufacturer "CPH2"

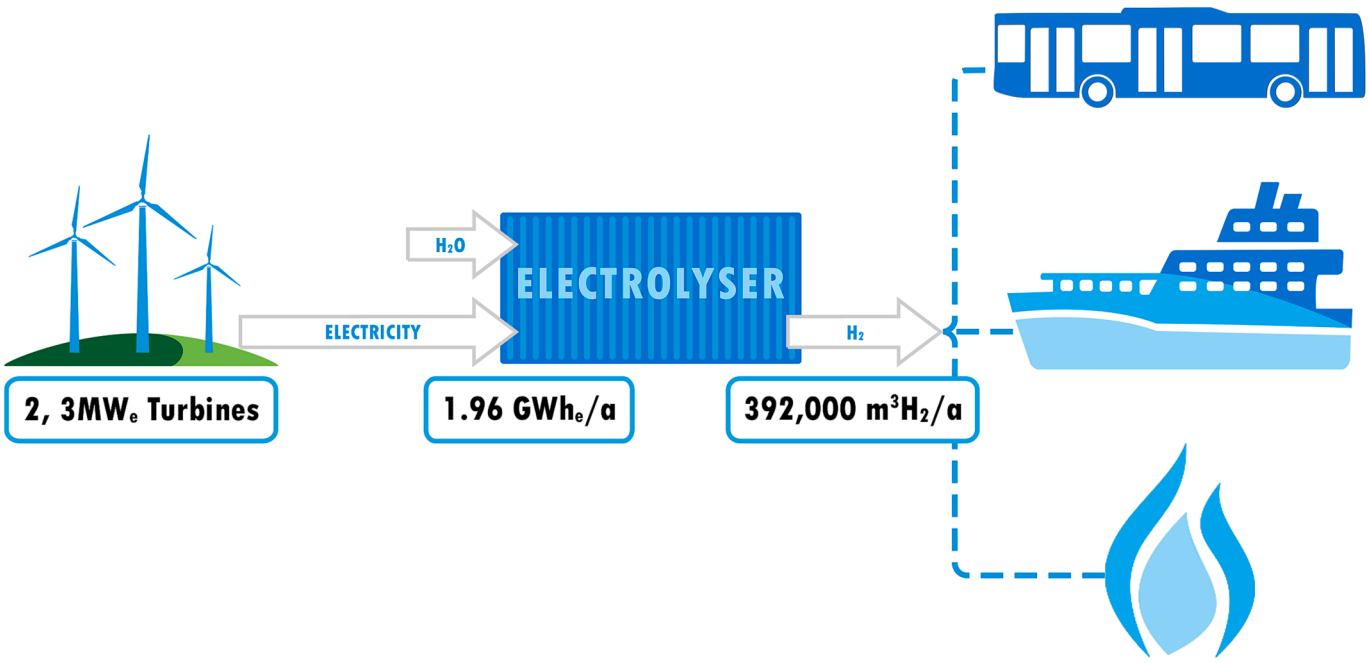


Fig. 2. Utilisation of all surplus VRE to produce green hydrogen for potential gas grid injection or use as a transport fuel. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

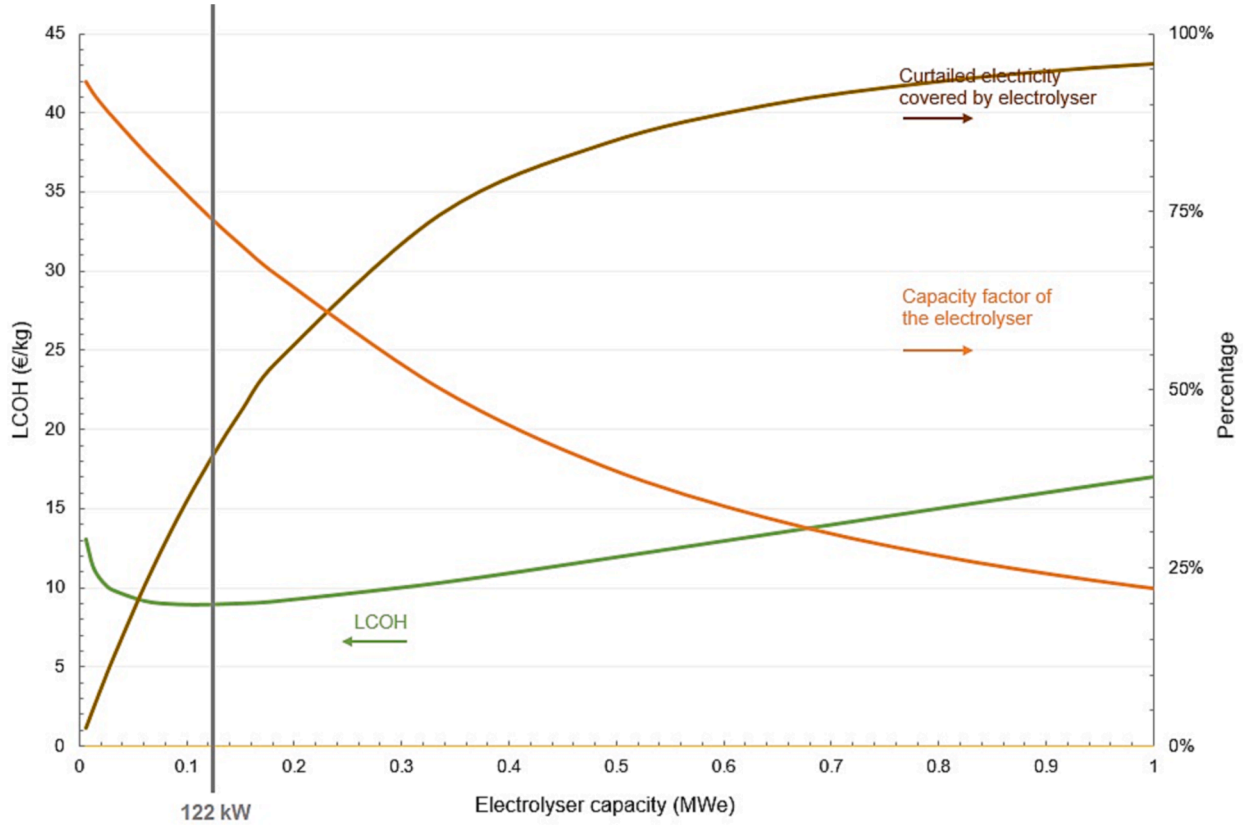


Fig. 3. Electrolyser capacity and levelized cost of hydrogen. The optimal 122 kW system is represented by a vertical black line intersecting the Capacity Factor curve (orange), the Curtailed Energy Capture curve (brown) and the LCOH curve (green). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

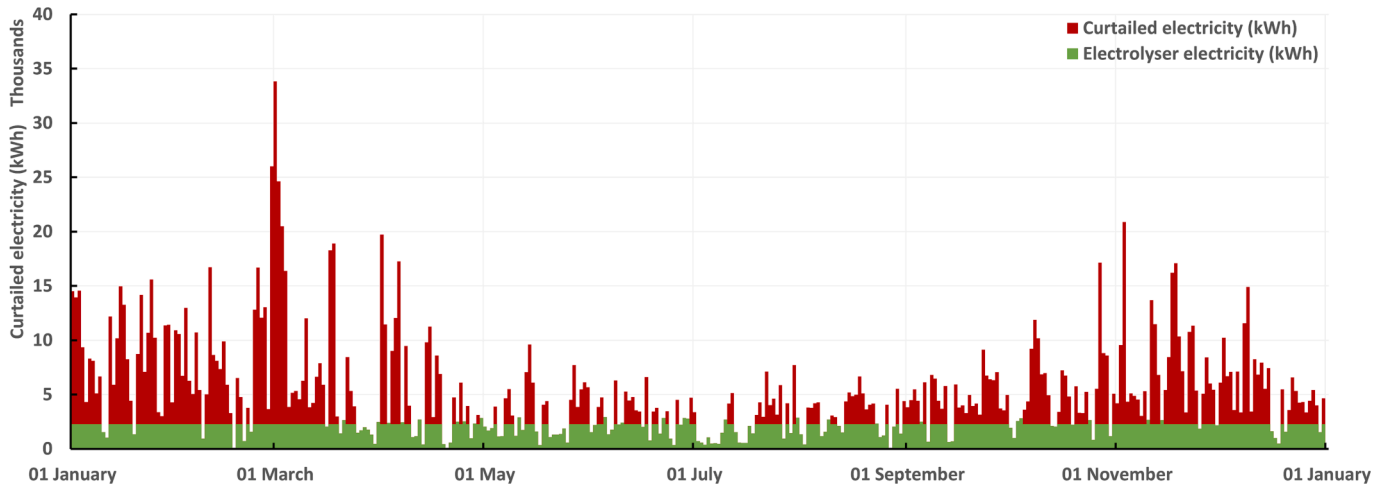


Fig. 4. Surplus electricity captured by the 122 kW PEM electrolyser at the industrial site.

with an investment cost of ca. €428,000.

3.3. Hydrogen as a transport fuel

Hydrogen is considered an advanced transport fuel and is particularly suitable for long-distance and heavy-goods transportation [4]. To facilitate a renewable transport sector, it was also proposed that the industry-derived hydrogen resource could be applicable to the local public bus service, or a commuter ferry given the specific industrial site is located in a harbour. An analysis of the associated hydrogen resource of 17,100 kgH_2/a from the industry site suggests the potential to provide the fuel necessary for 5 hydrogen buses providing 4 round trips per day from the industry site to Cork City (38 km round trip); or providing the fuel for 5,512 km of travel for a hydrogen commuter ferry with an estimated capacity of 84 passengers. The full calculations are shown in Box S2 in supplementary data. Such an approach would provide a means for zero-emissions transport from the proposed industry site to the local zone of major population.

The evidence presented in Box S2 in supplementary data suggests that the quantity of excess electricity is inherently small and most likely insufficient to run one ferry. The electrolyser that would be coupled with these turbines is also relatively small and this exacerbates the cost of hydrogen. The total cost of hydrogen for grid injection (which may be the most likely means of deployment) equates to approximately €10/kg (or €0.3/kWh or €3/L diesel equivalent) when including the cost of production and transportation. This can be compared to an analysis by McDonagh et al. [52] which suggested that the cost of hydrogen needs to be in the order of €4/kg H_2 to be financially viable from offshore wind (in a hybrid model producing electricity and hydrogen) as opposed to curtailment. It is also far above the target cost for 2030 set by the Hydrogen Council of approximately $2.50/kg (equivalent to €2.22/kg at a USD/EUR exchange rate of 0.89) [53].

The key learning from this analysis is that curtailed electricity from two number 3-MW wind turbines on its own would not be sufficient to make hydrogen financially competitive as a transport fuel. This is in agreement with other studies which suggest curtailed electricity alone may not be sufficient for cost effective hydrogen production and increasing operating hours by purchasing cheap electricity from the grid would be a critical function in making green hydrogen cost-effective [8,54]. In essence, scale can be considered the key enabler for green hydrogen production and large resources of electricity are required to run electrolysers cost-effectively.

3.4. Oxygen recycling to the wastewater treatment process

The total oxygen produced was calculated to be 135,660 kgO_2/a as per Box S3 in the supplementary data. Based on the influent wastewater properties (see Table S1 in Box S3 in the supplementary data), this oxygen resource accounts for only 8.9% of the estimated demand of the local WWTP in proximity to the industrial site which was sized at a capacity of 65,000 PE.

At WWTPs, fine-bubble diffusers are generally used to diffuse oxygen into the wastewater by pumping air. Transfer rates of 2.0–2.5 kg O_2/kWh$_{el}$ are typically achieved with this standard method [55]. The use of pure oxygen (from electrolysis) can reduce the on-site energy consumption associated with air compression as only approximately one-fifth of the volume is now required [25]. The study by Mohammadpour et al. [34] reported a 550% performance improvement with identical equipment, only by substituting pure O_2 gas in the aeration process, instead of air. This was due to factors such as high solubility, 500% higher O_2 driving force, and a 20% increase in the volumetric mass transfer coefficient for the smaller bubbles (relative to air). Therefore, if the pure oxygen resource from the 122 kW electrolyser (135,660 kg O_2/a) was fully utilised for aeration at the WWTP of 65,000 PE, the electricity saved to transfer oxygen demand would equate to 49.4 MWh$_{el}$/a (see Box S3 in supplementary data). Consequently, CO_2 emissions savings of approximately 16.04 tCO_2eq/a, or an overall reduction of 3.6% could be achieved through a reduction in electricity usage at the WWTP (see Box S3 in supplementary data). This analysis assumes that a pure oxygen stream from the electrolyser is added to the aeration system while the existing aeration system utilises air.

3.5. Biomethanation and the potential for carbon capture

Biomethanation converts CO_2 to CH_4 through the addition of H_2 in a Sabatier reaction (see Equation (8). In consideration of this case study, digestion of sewage sludge at the local WWTP with a design capacity of 65,000 PE has the potential to produce 214,800 m^3 CH_4/a and 118,200 m^3 CO_2/a (see Box S4 in supplementary data). Assuming that all of the CO_2 is captured and converted, the CH_4 output from the digester at the local WWTP could be increased by 53.8% (converting 118,200 m^3 CO_2 to CH_4 as per Box S4 in supplementary data). However, due to the relatively small electrolyser size (122 kW), there is not enough H_2 produced to capture all of the CO_2 in the biogas from the sludge digester. In the current scenario, the H_2 produced from a 122 kW electrolyser could convert 40% of the total CO_2 produced through a biomethanation process (Box S4 in supplementary data). Thus, in this instance, it may be more advantageous for the produced H_2 to be used directly as an

advanced transport fuel to decarbonise heavy transport or injected to the local gas grid network as described in Section 2.3. However larger scale electrolysers would again enable the potential for carbon capture at the WWTP.

In theory, hydrogen generated from electrolysis could capture CO_2 from any nearby digestion system (not exclusive to WWTP) and could allow for the specific targeting of hard-to-abate sectors such as agriculture. In Ireland, agriculture was responsible for 37.1 % of total GHG emissions in 2020, in which manure management contributed up to 10.3% of these emissions [56]. Hydrogen may potentially be used to capture the CO_2 from agricultural digesters treating slurries and manures to produce biomethane. In this way, hydrogen can offset the requirement for traditional biogas upgrading and the associated costs of such methods. Furthermore, any industry process generating CO_2 emissions would also be suitable for biomethanation, especially, large CO_2 stationary sources including ethanol fermentation plants (nearly 100% CO_2 by vol.) and cement manufacturers (average 20% CO_2 by vol.) where concentrated CO_2 streams exist [57]. This approach is further justified by the inherent corporate social responsibility of such industries in progressing towards carbon neutrality.

Fig. 5 presents a visual summary of the results obtained from the electrofuel in a circular economy system located at the WWTP.

3.6. Co-locating electrolysers and biomethanation at municipal wastewater treatment plants

WWTPs are usually located in urban locations offering geographically favourable site conditions to produce electrofuels with significant synergies in the coupled sectors (water, gas, electricity). Storage of hydrogen is expensive, but WWTPs with existing AD systems can use hydrogen to upgrade biogas. Co-locating electrolysers and biological methanation systems at existing WWTPs offers numerous benefits such as storage of intermittent renewable energy, decarbonisation of aeration processes at WWTPs with recycled O_2, the offset of traditional biogas upgrading costs (through carbon capture by biological methanation), and decarbonisation of heavy transport through advanced hydrogen, methane, or methanol production. Such developments to date have been scarce. However, the "Electrochaea BioCat" plant located at the local WWTP of 350,000 PE in Avedøre, Denmark demonstrated such novel technology. An on-site 600 kW AEC electrolyser was powered by renewable electricity in conjunction with the local power grid to generate hydrogen which was delivered to an on-site biological methanation reactor. CO_2 was sourced from biogas produced from a biogas plant in the local municipality [15]. Hydrogen and biogas were introduced to the biological methanation system at a stoichiometric ratio of 4:1 (H_2: CO_2) for further conversion to methane. The generated biomethane (which achieved a purity of 97 – 98.6%) was added to the local gas distribution grid, which operates at a pressure of 3 bar [17].

When including for biological methanation, the generated biomethane can be injected into the existing gas grid, allowing for easier transportation, distribution, and utilisation (compared to hydrogen). Biomethane can serve as a low-carbon transport fuel for buses and haulage. Alternatively, biomethane can be further synthesised to methanol. Methanol can be utilised in-situ as an exogenous carbon source in municipal WWTPs for heterotrophic denitrification to achieve low total nitrogen levels, presenting further circularity and enhancing overall the sustainability of the WWTP [58]. Methanol synthesis could also offer a potential solution in the maritime sector where it has been identified as a suitable fuel at a lower investment cost due to limited engine or bunkering modifications of current marine engines and easier storage features [4]. Biomethane and methanol as energy carriers can be used as interim solutions to a future green hydrogen economy. WWTPs can accompany that transformation by providing the currently required type of fuel and reducing the construction of interim solutions.

Overall, the potential for electrofuel production in a circular economy approach offers synergies in three key service sectors: electricity, gas, and water sectors, with numerous benefits such as maximising the use of the existing infrastructure with only minor modification and enabling the decarbonisation of hard-to-abate sectors (transport, wastewater treatment, industry, and agriculture).

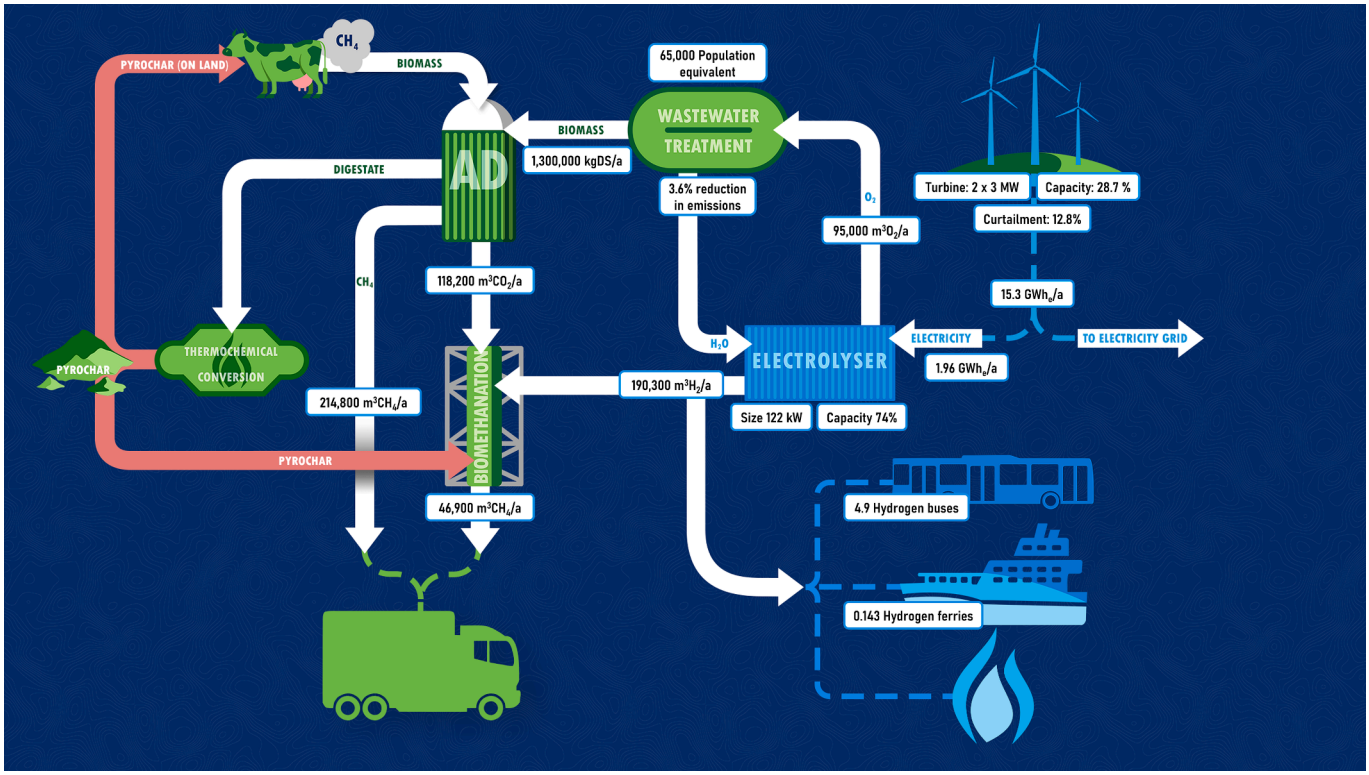


Fig. 5. Summary flow diagram of the circular economy system at a local industrial site.

3.7. Emission reductions using anaerobic digestion and pyrolysis

Pyrolysis of the digestate (derived from the AD system) to produce biochar has been identified as a potential NET. To assess such an integrated system, a case study was undertaken based on the available feedstock (cattle slurry, food waste and sewage sludge) within the 10 km area surrounding the investigated industrial site. Table 3 shows the potential GHG emissions reduction for this area through the proposed circular cascading system. When considering cattle slurry for AD, the fugitive emissions avoided from conventional slurry storage ought to be included in the GHG emissions savings; according to the recast Renewable Energy Directive (REDII), this equates to avoided emissions of 54kgCO_{2e}/t wet weight of slurry [59]. The results show that cattle slurry would comprise the majority of the GHG emissions savings through biogas production and upgrading via biomethanation, and biochar production via pyrolysis, totalling 35.25 ktCO_2 per year (see calculations in Box S5 of supplementary data). If all of the H_2 produced is used for biomethanation, ca. 46,900 m^3 CH_4 would be produced, equating to 234 tCO_2 in GHG emissions reductions. However, this represents only 1% of the total CO_2 in the biogas produced from the digested biomass within the 10 km area, due to the limited H_2 production. In effect, to utilise the remaining biogenic CO_2 (99% of the total CO_2), H_2 availability would be necessary in the form of a much larger electrolyser unit. Combining the abovementioned feedstocks, the integrated AD and pyrolysis system could potentially reduce emissions in total by 42.7 ktCO_2 per annum via biomethane and biochar production (Table 3).

3.8. The sustainability of the circular electrofuel production systems

The proposed methodology for a circular electrofuel production system integrates multiple processes, with the aim of not only deriving as much energy as possible but also maximising the potential for decarbonisation in the hard-to-abate sectors. Such an approach aligns with the European Green Deal policy proposing a multi-sectoral ambition to decarbonise. However, from an emissions accountancy perspective, some of the key circular attributes of this proposed system of systems are not accounted for.

The main technologies deployed are the electrolyser for generating H_2 and O_2 from surplus electricity; the AD plant for converting organic biomass to biogas; the biological methanation process for converting CO_2 and H_2 to CH_4; and pyrolysis of digestate to produce biochar for CO_2 sequestration. These systems are further integrated with other technologies such as wind turbines at the industrial site and the local wastewater treatment facility. Along with these technologies, the AD feedstock used (for example wastewater sludges, slurries or food wastes) will alter the sustainability of the circular system. For any electrofuel developed (H_2 or CH_4), it will be necessary to meet EU sustainability criteria under the recast Renewable Energy Directive to be certified as renewable [59]. A range of inputs, outputs and processes need to be considered in assessing the circular electrofuel system. Table 4 illustrates a list of initial considerations.

The RED recast has a sustainability criterion of a minimum of 70% GHG emissions savings for transport and 80% for heat by 2030 [59]. The criteria to be met for transport are more lenient than for heat due to a higher fossil fuel comparator and a lower requirement for GHG savings [63]. This highlights the exergy of the energy vector; wood chips can provide heat but not directly power a vehicle while biomethane or H_2 can do both. It is preferable to use a vector suitable for the hard to decarbonise sectors in the hard to decarbonise sector. Thus, the recast renewable energy directive (REDII) does not make it easy to satisfy the sustainability criteria for heat. Furthermore, electrofuels are typically aligned with a transport end use. However, it is evident that aspects of the circular approach presented do not currently benefit from a sustainability accounting process. For instance, whilst the biomethanation system will utilise the biomass derived CO_2 from AD, this is not considered carbon capture according to the recast REDII, and subsequently is not considered an emissions saving. Furthermore, the recast

Table 4
REDII Sustainability considerations for an electrofuel in a circular economy system.

Input	• Residues with no associated emissions (food waste, WWTP sludge) Renewable electricity (surplus VRE)
Processes	• Anaerobic digestion Transportation (feedstock and/or gas) Wastewater treatment plant Electrolysis Biological hydrogen methanation Compression of H_2/CH_4 Pyrolysis
Output	• Transport fuel (direct use) Heat (direct use) Grid injection
Sustainability Criteria	• Recast REDII 70% emissions savings for transport fuel as compared to fossil fuel comparator 80% emissions savings for heat as compared to fossil fuel comparator

Table 3
Potential GHG reduction by the proposed circular system. (The quantification of available feedstock in the 10 km area surrounding the industrial area was based on a previous study [60]).

Feedstock	Resource (kt, fresh matter) [a]	Biogas production potential ($\times 10^3$ m^3)	Biomethane potential ($\times 10^3$ m^3)[b]	GHG reduction from use of CH_4 ($ktCO_2$)[c]	Avoided methane emissions potential ($ktCO_2$)	GHG reduction from biochar ($ktCO_2$)[d]	Total GHG reduction ($ktCO_2$)[e]
Cattle slurry	**267.93**	**7,127**	**4,287**	**11.31**	**14.47**	**9.46**	**35.25**
Food waste	**12.83**	**3,167**	**1,900**	**5.01**	/	**1.52**	**6.54**
Primary Sludge	**0.71 (dry matter)**	**282**	**189**	**0.50**	/	**0.19**	**0.69**
Waste Activated Sludge	**0.59 (dry matter)**	**49**	**25**	**0.07**	/	**0.16**	**0.22**
Total		**10,625**	**6,401**	**16.89**	**14.47**	**11.33**	**42.7**

[a] *: the TS and* VS *content of slurry and food waste is modelled at 8.75% and 6.69%* [61]*, and 29% and 28%* [62]*, respectively. The biomethane potential of cattle slurry is 16 m^3/t wet weight* [23]*, and food waste is 148 m^3/t wet weight* [62]*; the content of CH_4 and CO_2 are assumed as 60% and 40% respectively. The methane production potential and biogas composition of sewage sludge can be found in Box S5 in* supplementary data.

[b] *: Only including the methane production from the feedstock.*

[c] *: The use of methane (energy content of 10 kWh/m^3 CH_4) is modelled as a replacement of diesel consumption; the CO_2 emission factor of diesel is 263.9 g CO_2/kWh.*

[d] *: Biochar yield from dry matter of cattle slurry, food waste and sewage sludge (including primary sludge and waste-activated sludge) is conservatively assumed as 20%, 20% and 40%, respectively; the carbon content of the separate biochar is 55%, 55% and 18%, respectively* [48,50].

[e] *: Including the replacement of diesel consumption by biomethane and the calculation in Box S2 in* Supplementary data.

REDII provides no clear guidance for calculating emissions from surplus electricity that would otherwise not be used, which is an increasing issue in a growing VRE energy landscape. These accounting blind spots result in significant hurdles in the efficacy of decarbonising more difficult sectors, and ultimately hinder achieving net-zero goals.

A full life cycle assessment (LCA) would be required to assess the benefits of individual modular systems, while also providing an accurate assessment of the emissions savings such a system could offer to the surrounding industry and the hard-to-abate sectors. As this study has shown, the utilisation of curtailed VRE could offset the use of emission-intense fuels through electrolysis. The use of O_2 could offer reductions in total WWTP electricity demand. Additionally, the by-product CO_2 could be upgraded using the biological methanation process to a low-carbon biomethane when coupled with green H_2 ($4H_2 + CO_2 = CH_4 + 2H_2O$); these energy vectors could be used in the decarbonisation of the hard-to-abate sectors. The inclusion of pyrolysis-derived biochar can affect further emissions savings and improve the ability to comply with the REDII recast sustainability criteria to ensure the progression towards a net-zero economy.

3.9. Study limitations and future research

Whilst this study provided a high-level overview for a circular electrofuel production concept, it must be acknowledged that there are limitations to the analysis which may lead to additional questions and the potential for future research. For example, the study can be considered somewhat low-resolution as it focuses primarily on the movement of resources throughout the system. To effectively develop a higher resolution system, it is important that all aspects of the system are developed at a high level of detail. This could include for further analyses of the wastewater treatment process including for detailed designs of the aeration process, sizing of the aeration tanks and the exact influent properties, to determine the oxygen gas transfer efficiency. Furthermore, the wind resource could be assessed on a demand versus production basis, to estimate the periods of availability of the electricity. The investigation of a larger electrolyser size is warranted along with a cost-benefit analysis to determine the economic viability of co-locating the proposed systems, along with a techno-economic analysis for the total cost of the system. Additionally, factors such as the conversion efficiency of specific AD feedstocks and subsequent biogas yields would need to be accurately determined. With respect to the biomethanation process system, conversion efficiency and rate would need to be determined to evaluate the system ability to upgrade the CO_2 and furthermore the fugitive emissions of the process would need to be established. Nonetheless the system proposed herein provides a valuable roadmap for such technology integration to reduce emissions on a multi-sectoral level. Future research could investigate the integration of the technologies proposed into other industry fields such as distilleries or cement manufacture which offer a CO_2 source for biomethanation and where the products (hydrogen, methane and oxygen) could be utilised favourably.

4. Conclusions

An industrial site was investigated for its potential to store variable renewable electricity in the form of green hydrogen using an electrolyser. Based on the site data, a 122 kW proton exchange membrane electrolyser was sized to account for the surplus wind resource available. Recycling of the oxygen generated in electrolysis could supply 8.9% of the oxygen demand for the aeration process of the local 65,000 person equivalent wastewater treatment plant and reduce emissions by 16.04 tCO_2eq/a. The hydrogen produced from the electrolyser (190,300 m^3H_2/a) could be best used directly as a transport fuel in local buses or could convert 40% of CO_2 generated in anaerobic digestion at the wastewater treatment plant to biomethane in a biomethanation process. The levelized cost of hydrogen production was found to be high at €8.92/kg H_2. Thus, smaller scale systems using intermittent curtailed electricity as described can be deemed difficult to implement. However, larger scale systems with higher capacity factors stemming from more available electricity sources may be more financially competitive and offer significant decarbonisation opportunities. An integrated anaerobic digestion and pyrolysis system was found to potentially reduce emissions by 42.7 $ktCO_2/a$ via biomethane and biochar production (as a means for long-term CO_2 sequestration). The proposed circular electrofuel system can tackle the hard-to-abate sectors however the current emissions accountancy process may ultimately hinder the development of such approaches.

CRediT authorship contribution statement

Davis Rusmanis: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft. **Yan Yang:** Writing – original draft, Methodology. **Aoife Long:** Methodology, Data curation. **Nathan Gray:** Methodology, Data curation. **Kelvin C. Martins:** Methodology, Data curation. **Seán Óg Ó Loideáin:** Methodology, Data curation. **Richen Lin:** Methodology, Data curation. **Xihui Kang:** Methodology, Data curation. **Donal Óg Cusack:** Data curation. **James G. Carton:** Methodology, Data curation. **Rory Monaghan:** Methodology, Data curation. **Jerry D. Murphy:** Conceptualization, Methodology, Writing – review & editing, Supervision, Validation, Funding acquisition. **David M. Wall:** Conceptualization, Methodology, Writing – review & editing, Supervision, Validation, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.enconman.2023.117367.

References

[1] U. N. F. C. on C. C. UNFCCC,. “Conference of the Parties. Adoption of the Paris Agreement” 2015.
[2] Commission E. The European Green Deal. Eur Comm 2019;53(9):24. https://doi.org/10.1017/CBO9781107415324.004.
[3] SEAI, “Energy in Ireland,. Online. Available 2022”, 2022.. https://www.seai.ie/publications/Energy-in-Ireland-2020.pdf.
[4] Gray N, McDonagh S, O’Shea R, Smyth B, Murphy JD. Decarbonising ships, planes and trucks: An analysis of suitable low-carbon fuels for the maritime, aviation and haulage sectors. Adv Appl Energy 2021;1:100008. https://doi.org/10.1016/j.adapen.2021.100008.
[5] C. and C. Department of the Environment, “Climate Action Plan 2021: Tackling Climate Breakdown,” 2021. [Online]. Available: https://steel.gov.in/sites/default/files/Annual Report-Ministry of Steel 2020-21.pdf.

[6] EirGrid and SONI, "Annual Renewable Energy Constraint and Curtailment Report 2020," 2021. Accessed: May 26, 2021. [Online]. Available: https://www.soni.ltd.uk/media/documents/Annual-Renewable-Constraint-and-Curtailment-Report-2020-V1.0.pdf.

[7] Lehtveer M, Brynolf S, Grahn M. What Future for Electrofuels in Transport? Analysis of Cost Competitiveness in Global Climate Mitigation. Env Sci Technol 2019;53(3):1690–7. https://doi.org/10.1021/acs.est.8b05243.

[8] McDonagh S, O'Shea R, Wall DM, Deane JP, Murphy JD. Modelling of a power-to-gas system to predict the levelised cost of energy of an advanced renewable gaseous transport fuel. Appl Energy 2018;215(January):444–56. https://doi.org/10.1016/j.apenergy.2018.02.019.

[9] Ababneh H, Hameed BH. Electrofuels as emerging new green alternative fuel: A review of recent literature. Energy Convers Manag 2022;254:115213. https://doi.org/10.1016/J.ENCONMAN.2022.115213.

[10] Thema M, Bauer F, Sterner M. Power-to-Gas: Electrolysis and methanation status review. Renew Sustain Energy Rev 2019;112:775–87. https://doi.org/10.1016/j.rser.2019.06.030.

[11] Jones MP, Krexner T, Bismarck A. Repurposing Fischer-Tropsch and natural gas as bridging technologies for the energy revolution. Energy Convers Manag 2022;267:115882. https://doi.org/10.1016/J.ENCONMAN.2022.115882.

[12] Pascuzzi S, Anifantis AS, Blanco I, Mugnozza GS. Electrolyzer performance analysis of an integrated hydrogen power system for greenhouse heating a case study. Sustain 2016;8(7):1–15. https://doi.org/10.3390/su8070629.

[13] Capodaglio AG, Olsson G. Energy issues in sustainable urban wastewater management: Use, demand reduction and recovery in the urban water cycle. Sustain 2020;12(1):266. https://doi.org/10.3390/su12010266.

[14] Rusmanis D, Yang Y, Lin R, Wall DM, Murphy JD. Operation of a circular economy, energy, environmental system at a wastewater treatment plant. Adv Appl Energy 2022;8:100109. https://doi.org/10.1016/J.ADAPEN.2022.100109.

[15] D. T. Laurent Lardon Lars Krosgaard, "Technical and economic analysis of biological methanation," 2020. [Online]. Available: http://www.powerstep.eu.

[16] Appels L, Baeyens J, Degrève J, Dewil R. Principles and potential of the anaerobic digestion of waste-activated sludge. Prog Energy Combust Sci 2008;34(6):755–81. https://doi.org/10.1016/J.PECS.2008.06.002.

[17] J. Liebetrau P. Kornatz, U. Baier, D. Wall, and J. D. Murphy., "Integration of Biogas Systems into the Energy System: Technical aspects of flexible plant operation," IEA Bioenergy Task 37, 2020.

[18] Jacob, "Jacobs Yarra valley water towards a zero carbon future," 2020. https://www.jacobs.com/sites/default/files/2020-06/jacobs-yarra-valley-water-towards-a-zero-carbon-future.pdf.

[19] N. Gray, R. O'Shea, B. Smyth, P. N. L. Lens, and J. D. Murphy, "What is the energy balance of electrofuels produced through power-to-fuel integration with biogas facilities?," *Renew. Sustain. Energy Rev.*, p. 111886, Nov. 2021, doi: 10.1016/j.rser.2021.111886.

[20] Lund H, et al. The role of sustainable bioenergy in a fully decarbonised society. Renew Energy 2022;196:195–203. https://doi.org/10.1016/j.renene.2022.06.026.

[21] Ruggles TH, Dowling JA, Lewis NS, Caldeira K. Opportunities for flexible electricity loads such as hydrogen production from curtailed generation. Adv Appl Energy 2021;3:100051. https://doi.org/10.1016/J.ADAPEN.2021.100051.

[22] Wu B, Lin R, O'Shea R, Deng C, Rajendran K, Murphy JD. Production of advanced fuels through integration of biological, thermo-chemical and power to gas technologies in a circular cascading bio-based system. Renew Sustain Energy Rev 2021;135:110371. https://doi.org/10.1016/j.rser.2020.110371.

[23] Lin R, O'Shea R, Deng C, Wu B, Murphy JD. A perspective on the efficacy of green gas production via integration of technologies in novel cascading circular bio-systems. Renew Sustain Energy Rev 2021;150:111427. https://doi.org/10.1016/j.rser.2021.111427.

[24] Gao Y, Gao X, Zhang X. The 2 °C Global Temperature Target and the Evolution of the Long-Term Goal of Addressing Climate Change—From the United Nations Framework Convention on Climate Change to the Paris Agreement. Engineering 2017;3(2):272–8. https://doi.org/10.1016/J.ENG.2017.01.022.

[25] M. Schäfer, O. Gretzschel, and H. Steinmetz, "The Possible Roles of Wastewater Treatment Plants in Sector Coupling," *Energies*, vol. 13, no. 8, 2020, doi: 10.3390/en13082088.

[26] Brynolf S, Taljegard M, Grahn M, Hansson J. Electrofuels for the transport sector: A review of production costs. Renew Sustain Energy Rev Jan. 2018;81:1887–905. https://doi.org/10.1016/j.rser.2017.05.288.

[27] European Commission, "Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions - A hydrogen strategy for a climate-neutral Europe," 2020.

[28] Buttler A, Spliethoff H. Current status of water electrolysis for energy storage, grid balancing and sector coupling via power-to-gas and power-to-liquids: A review. Renew Sustain Energy Rev 2018;82:2440–54. https://doi.org/10.1016/j.rser.2017.09.003.

[29] Burton NA, Padilla RV, Rose A, Habibullah H. Increasing the efficiency of hydrogen production from solar powered water electrolysis. Renew Sustain Energy Rev Jan. 2021;135:110255. https://doi.org/10.1016/j.rser.2020.110255.

[30] Gunawan TA, Singlitico A, Blount P, Burchill J, Carton JG, Monaghan RFD. At what cost can renewable hydrogen offset fossil fuel use in Ireland's gas network? Energies Apr. 2020;13(7):1798. https://doi.org/10.3390/en13071798.

[31] Rusmanis D, O'Shea R, Wall DM, Murphy JD. Biological hydrogen methanation systems – an overview of design and efficiency. Bioengineered Jan. 2019;10(1):604–34. https://doi.org/10.1080/21655979.2019.1684607.

[32] GNI, "Vision 2050 A Net Zero Carbon Gas Network for Ireland," 2019.

[33] Schemme S, Samsun RC, Peters R, Stolten D. Power-to-fuel as a key to sustainable transport systems - An analysis of diesel fuels produced from CO2 and renewable electricity. Fuel 2017;205:198–221. https://doi.org/10.1016/j.fuel.2017.05.061.

[34] Mohammadpour H, Cord-Ruwisch R, Pivrikas A, Ho G. Utilisation of oxygen from water electrolysis – Assessment for wastewater treatment and aquaculture. Chem Eng Sci Dec. 2021;246:117008. https://doi.org/10.1016/J.CES.2021.117008.

[35] I. Environmental Protection Agency, "Wastewater Treatment Manuals," 1997.

[36] Wei W, et al. Zero valent iron enhances methane production from primary sludge in anaerobic digestion. Chem Eng J Nov. 2018;351:1159–65. https://doi.org/10.1016/J.CEJ.2018.06.160.

[37] Wei W, et al. Enhanced high-quality biomethane production from anaerobic digestion of primary sludge by corn stover biochar. Bioresour Technol Jun. 2020;306:123159. https://doi.org/10.1016/J.BIORTECH.2020.123159.

[38] Potdukhe RM, Sahu N, Kapley A, Kumar R. Co-digestion of waste activated sludge and agricultural straw waste for enhanced biogas production. Bioresour Technol Reports Sep. 2021;15:100769. https://doi.org/10.1016/j.biteb.2021.100769.

[39] EASAC, "EASAC report on negative emission report," 2018. doi: https://easac.eu/publications/details/easac_net/.

[40] Guilayn F, Rouez M, Crest M, Patureau D, Jimenez J. Valorization of digestates from urban or centralized biogas plants: a critical review. Rev Environ Sci Bio/Technology Jun. 2020;19(2):419–62. https://doi.org/10.1007/s11157-020-09531-3.

[41] Deng C, Lin R, Kang X, Wu B, O'Shea R, Murphy JD. Improving gaseous biofuel yield from seaweed through a cascading circular bioenergy system integrating anaerobic digestion and pyrolysis. Renew Sustain Energy Rev 2020;128:109895. https://doi.org/10.1016/j.rser.2020.109895.

[42] Spokas KA. Review of the stability of biochar in soils: predictability of O: C molar ratios. Carbon Manag 2010;1(2):289–303.

[43] Lin R, Cheng J, Ding L, Murphy JD. Improved efficiency of anaerobic digestion through direct interspecies electron transfer at mesophilic and thermophilic temperature ranges. Chem Eng J 2018;350:681–91. https://doi.org/10.1016/j.cej.2018.05.173.

[44] Wu B, et al. Graphene Addition to Digestion of Thin Stillage Can Alleviate Acidic Shock and Improve Biomethane Production. ACS Sustain Chem Eng Sep. 2020;8(35):13248–60. https://doi.org/10.1021/acssuschemeng.0c03484.

[45] Li C, Zhu X, Angelidaki I. Carbon monoxide conversion and syngas biomethanation mediated by different microbial consortia. Bioresour Technol 2020;314:123739.

[46] O'Shea R, Wall DM, Kilgallon I, Browne JD, Murphy JD. Assessing the total theoretical, and financially viable, resource of biomethane for injection to a natural gas network in a region. Appl Energy Feb. 2017;188:237–56. https://doi.org/10.1016/j.apenergy.2016.11.121.

[47] Voelklein MA, R. O' Shea, A. Jacob, and J. D. Murphy,. Role of trace elements in single and two-stage digestion of food waste at high organic loading rates. Energy 2017;121:185–92. https://doi.org/10.1016/j.energy.2017.01.009.

[48] Zielińska A, Oleszczuk P, Charmas B, Skubiszewska-Zięba J, Pasieczna-Patkowska S. Effect of sewage sludge properties on the biochar characteristic. J Anal Appl Pyrolysis 2015;112:201–13.

[49] Callegari A, Capodaglio A. Properties and Beneficial Uses of (Bio)Chars, with Special Attention to Products from Sewage Sludge Pyrolysis. Resources Mar. 2018;7(1):20. https://doi.org/10.3390/resources7010020.

[50] Monlau F, Sambusiti C, Ficara E, Aboulkas A, Barakat A, Carrère H. New opportunities for agricultural digestate valorization: current situation and perspectives. Energy Environ Sci 2015;8(9):2600–21. https://doi.org/10.1039/c5ee01633a.

[51] Ding Y, et al. Biochar to improve soil fertility. A review. Agron Sustain Dev 2016;36(2):1–18.

[52] McDonagh S, Ahmed S, Desmond C, Murphy JD. Hydrogen from offshore wind: Investor perspective on the profitability of a hybrid system including for curtailment. Appl Energy May 2020;265:114732. https://doi.org/10.1016/j.apenergy.2020.114732.

[53] H. Council, "Path to hydrogen competitiveness A cost perspective," 2020. https://hydrogencouncil.com/en/path-to-hydrogen-competitiveness-a-cost-perspective/.

[54] McDonagh S, Wall DM, Deane P, Murphy JD. The effect of electricity markets, and renewable electricity penetration, on the levelised cost of energy of an advanced electro-fuel system incorporating carbon capture and utilisation. Renew Energy Feb. 2019;131:364–71. https://doi.org/10.1016/j.renene.2018.07.058.

[55] Drewnowski J, Remiszewska-Skwarek A, Duda S, Łagód G. "Aeration Process in Bioreactors as the Main Energy Consumer in a Wastewater Treatment Plant. Review of Solutions and Methods of Process Optimization", *Processes* May 2019;7(5):311. https://doi.org/10.3390/pr7050311.

[56] EPA, "Greenhouse gas emissions Ireland," 2019. https://www.epa.ie/ghg/agriculture/.

[57] John Bradshaw *et al.*, "Sources of CO2," J. F. H. Ismail El Gizouli, Ed. 2018.

[58] Su YC, Sathyamoorthy S, Chandran K. Bioaugmented methanol production using ammonia oxidizing bacteria in a continuous flow process. Bioresour Technol 2019;279:101–7. https://doi.org/10.1016/j.biortech.2019.01.092.

[59] EU Directive, "Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on the promotion of the use of energy from renewable sources." [Online]. Available: http://data.europa.eu/eli/dir/2018/2001/oj.

[60] O'Shea R, Kilgallon I, Wall D, Murphy JD. Quantification and location of a renewable gas industry based on digestion of wastes in Ireland. Appl Energy 2016;175(2016):229–39. https://doi.org/10.1016/j.apenergy.2016.05.011.

[61] Wall DM, O'Kiely P, Murphy JD. The potential for biomethane from grass and slurry to satisfy renewable energy targets. Bioresour Technol 2013;149:425–31.
[62] Browne JD, Murphy JD. Assessment of the resource associated with biomethane from food waste. Appl Energy 2013;104:170–7.
[63] Long A, Bose A, O'Shea R, Monaghan R, Murphy JD. Implications of European Union recast Renewable Energy Directive sustainability criteria for renewable heat and transport: Case study of willow biomethane in Ireland. Renew Sustain Energy Rev 2021;150:111461. https://doi.org/10.1016/j.rser.2021.111461.