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Exploring the future of photosynthetic biogas upgrading process

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The increasing demand for biomethane in Europe requires the urgent development of sustainable and cost-effective technologies to meet the ambitious target of producing 35 bcm of biomethane by 2030. Biological technologies stand as a promising integrating platform to purify biogas and valorize biogenic CO_2, while producing high-value chemicals. Photosynthetic biogas upgrading has demonstrated to be an attractive platform as microalgae can fix CO_2 while producing biomass that can be further valorized for biofuel, biofertilizer or biostimulant production. However, its high investment cost and the low microalgae biomass productivities have limited commercialization. Recently, the supplementation of nanoparticles obtained from olive-mill wastewater, has demonstrated to be a promising technique to improve biomass productivity, CO_2 removals, and the overall stability of the system. This review summarizes the current trends and future outlooks of this sustainable platform aiming at the development of novel integrated microalgae biorefineries devoted to biogas upgrading and high-value compounds production.

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Introduction

The ambitious 2030 and 2050 environmental policies targeting net-zero emissions, in addition to the current war conflicts in Europe, have accelerated the use of renewables, particularly biogas [1].

Biogas produced via anaerobic digestion (AD) stands as a cost-effective energy vector mainly composed of CH_4 (65–70%, v/v) and CO_2 (45–30%, v/v), and other traces of contaminants including H_2S, O_2, N_2, and volatile organic compounds in minor concentrations. To improve biogas energy power, CO_2 and H_2S must be removed to generate a high-quality biomethane analogous to natural gas.

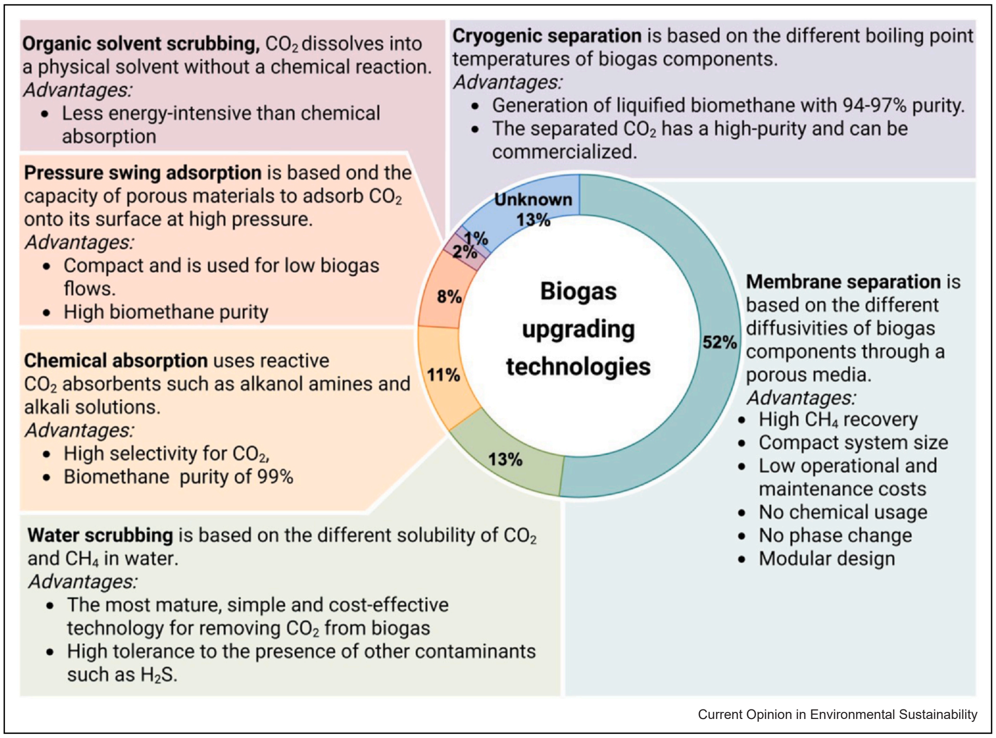
From the year 2023 to 2024, the European investments in the biogas market accounted for €345 million, and future investments of €28.4 billion for developments inside and outside Europe are foreseen by 2030 [2]. New plants producing biomethane will take 85% of the future investments, while the rest will be devoted to upgrading existing biogas-CHP plants to biomethane plants. This intense demand for biomethane plants fosters the urgent development of sustainable biogas upgrading technologies.

This work presents the current trends in biogas upgrading technologies with particular emphasis on photosynthetic biogas upgrading (PBU) process. The 50 peer-reviewed studies with the highest impact in the field, mainly from 2020 to 2025, are presented and discussed to support an equilibrated opinion regarding PBU. We first address the state-of-the-art of commercial biogas upgrading technologies, their position in the market, and the need for sustainable biogas upgrading technologies to meet the ambitious targets of the European Commission to produce 35 bcm of biomethane by 2030. Then, the trending biological biogas upgrading technologies are addressed, discussed, and compared to PBU. Thereafter, the fundamentals of PBU, process limitations, and opportunities to improve the system sustainability by relying on the use of nutrient-rich effluents are discussed. In addition, the current use of nanoparticle supplementation and its role in high-value compounds to improve PBU technoeconomic feasibility is addressed. Finally, the concluding remarks are presented.

Overview of biogas upgrading technologies and future market projections

Different physicochemical technologies are currently available in the market to upgrade biogas aiming to comply with international biomethane regulations for gas injection into the natural gas grid or use as autogas

Figure 1



Current biogas upgrading technologies commercially available and their market share in Europe during 2023 [3].

(Figure 1). Biomethane end-use will determine the quality of its composition, but in Europe, most countries are regulated by the EN 16723:2018, which requires a composition of CH_4 ≥90%, CO_2 ≤2%, O_2 ≤1% and a negligible concentration of H_2S for grid injection.

Typically, the upgrading process accounts for up to 40% of the total investment cost of a biomethane plant and 30% of the cost of biomethane, with CO_2 removal being the most expensive step, resulting in an average biomethane production cost between €9 and 25/GJ [2].

Sustainable and competitive upgrading technologies, namely biological upgrading technologies, can reduce biomethane production cost to €7/GJ. However, the coupled production of valuable by-products from biogas and digestate is crucial to increase biomethane competitiveness and circularity.

Current trends in biological biogas upgrading processes

In 2023, with the creation of the Biomethane Industrial Partnership, innovative upgrading technologies, namely, methanation (biological and chemical) and PBU, were projected as fundamental to accelerate the biomethane industry scale-up [3]. Despite biological technologies' great potential, their current high investment and operational costs will require additional government support and political regulations before commercialization [4]. Hence, a biorefinery approach to concomitantly produce biomethane and high-value chemicals along with wastewater/digestate treatment could potentially improve their commercialization.

Recently, three biological upgrading technologies could potentially gain market weight (Table 1). Undoubtedly, biological methanation (BM) represents a more feasible technology as it can be directly integrated into existing plants by injecting H_2 to the digester or in an external reactor. BM can reduce the need to install further upgrading operational units and can increase biomethane yield without generating any additional subproducts [5–10]. Recently, Jønson et al. [7] demonstrated the potential of ex-situ BM in a pilot-scale biogas plant. After testing different operation configurations, CH_4 yields between 9.4 and 10.6 $Nm^3_{CH4}/m^3_{Reactor}d$ with CH_4 contents between 95.7% and 97.4%, were obtained. The system demonstrated robustness and ability to recover and maintain high BM efficiencies after H_2 supply losses; nonetheless, one of the main bottlenecks to address is the cost-effective H_2 production [7]. Currently, H_2 is commercially produced by water electrolysis, a high-energy-demand process that relies on the use of fossil fuels, which is neither attractive nor competitive

Table 1

Comparison of fundamentals of the trending biological biogas upgrading technologies.

Biological upgrading technology	Methanation	Organic acid production	PBU
Technology fundamentals	Based on the conversion of H_2 and CO_2 to CH_4 according to the chemical equation: $CO_2 + 4H_2 \rightarrow CH_4 + H_2O$ Can be conducted via chemical and biological pathways External H_2 is used to convert CO_2 from biogas to CH_4	Based on the sugar fermentation and CO_2 capture from biogas with anaerobic bacteria, resulting in the production of organic acids.	Based on the use of microalgae-bacteria consortia to symbiotically remove CO_2, H_2S, NH_3, and other volatile organic compounds from biogas via metabolic pathways, mainly photosynthesis and nitrification
Microorganisms used	Hydrogenotrophic methanogenic archaea	Anaerobic bacteria such as *Actinobacillus succinogenes* 130Z, *Anaeribiospirillum succiniciproducens, Basfia succiniciproducens*, and *Mannheimia succiniciproducens*	Microalgae and nitrifying bacteria
System configurations	*In-situ*. Methanation is conducted in the existing anaerobic digester. Can be more cost-effective but bacterial competition within the reactor is higher. *Ex-situ*. Requires an external and independent reactor. Trickle-bed reactors have been widely used. More selective and can reach higher CO_2 conversion	Still in early stages of deployment, but requires an anaerobic fermenter.	Consists of a HRAP, where microalgae-bacteria biomass is grown, connected to an absorption column where biogas is sparged to be upgraded to biomethane. A settler is placed in between to maximize biomass production, and to allow the circulation of microalgae biomass from the HRAP to the column
Technology advantages	High CO_2 conversion In-situ methanation could reduce the need of external upgrading Enhanced biomethane yield	Production of valuable precursors for chemical industry No O_2 contamination	High-value products production Use of sunlight as energy Nutrient recycling from digestates
Technology drawbacks	Need for a costly hydrogen source Low mass-transfer of H_2 to microorganisms High H_2 partial pressure inside the reactor can inhibit hydrolysis and acetogenesis, resulting in VFA accumulation High sensitivity to alkaline pH	Low bacteria selectivity to inorganic carbon. High concentrations of costly sugars are required	The use of microalgae increases the risk of O_2 contamination in the upgraded biomethane. Land and water use High investment costs
Investment cost	CAPEX is around 20–200 €/MWh and OPEX 13 €/MWh (0.4 kWh electricity/Nm^3 biogas)	N.A.	Dependent on plant size, but an average plant. For instance a CAPEX of €6000/(Nm^3/h) was estimated for a biogas capacity of 300 Nm^3/h
Energy demand	It is estimated as the energy needed to produce H_2 via electrolysis, 43–66 kWh/kg_{H2}	It has been estimated as 2–19% of the total manufacturing costs	It is estimated as 22.8 kWh/d for a biogas plant producing 120 m^3_{biogas}/d
CH_4 purity	> 92%	*85%*	> 92%
Subproduct obtained/yield	CH_4 is the main product	Succinic acid is the targeted acid, yield of 0.92 g/$g_{substrate}$	Biomass, productivity up to 22 g/m^2d
Use of wastewater	Yes, centrate	Yes, potato wastewater	Yes, mainly centrate
References	[12,13,15,16,18,19]	[4–8]	[3,9–11,20]

N.A., not applicable.VFA, volatile fatty acids

[9]. Hence, other H_2 supplies route, including the use of renewable energy, must be developed to meet BM H_2 demand.

Conversely, the production of organic acids, namely succinic acid (SA), via CO_2 fermentation is emerging as a promising upgrading technology entailing a high purity biomethane > 90% [4,11–14]. Yet, this technology is still in its early stages and has not reached pilot-scale demonstration. Moreover, the low selectivity of anaerobic bacteria to inorganic carbon has been a major drawback for its implementation [15], particularly as high CO_2 removal efficiencies are targeted during biogas upgrading. Key research advances have been made to increase bacterial CO_2 affinity, namely $MgCO_3$ supplementation, while sustaining high SA yields of 0.6 g_{SA}/$g_{substrate.}$ However, the upgraded biomethane did not meet the EN 16723:2018 thresholds [15].

In this regard, PBU stands as a more feasible technology since non-expensive energy carriers to produce biomethane are needed. Microalgae robustness, ability to adapt to extreme climate conditions, and high affinity for CO_2, result in a biomethane that meets the EN 16723:2018 standard, highlighting the benefits and scalability of PBU. PBU is a more developed technology that has been extensively studied, optimized, and validated at pilot and demo-scale [16]. During PBU, the upgraded biomethane can be potentially contaminated with O_2, unlike the other biological technologies, but strategies to reduce O_2 in biomethane have been implemented to reduce O_2 to concentrations < 1%. To date, PBU has been limited to small-scale biogas plants mainly due to the large superficial areas needed for the photobioreactors. However, an improved microalgae metabolism and the coupled production of high-value chemicals would facilitate its implementation in medium and large-scale plants.

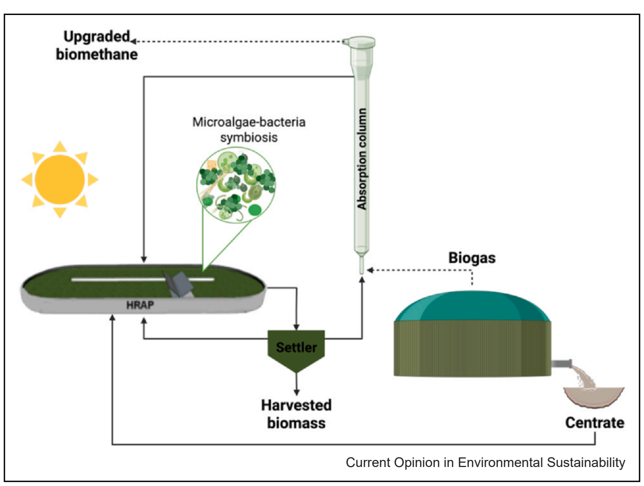
Photosynthetic biogas upgrading

Fundamentals of the process under a circular economy approach

PBU system configuration basically consists of a high rate algal pond (HRAP), where microalgae-bacteria biomass is grown, connected to an absorption column where biogas is sparged to be upgraded to biomethane (Figure 2).

During PBU, microalgae fix CO_2 from biogas via water photolysis and uptake nitrogen and phosphorus, via assimilatory mechanisms, in the so-called photosynthesis process, resulting in an excess of O_2 that promotes the oxidation of H_2S to SO_4^{2-} and NH_3 to NO_3^- by bacteria [9]. The intense demand of O_2 to remove contaminants from biogas results in a high-quality biomethane with

Figure 2



Schematic representation of the PBU system: a HRAP interconnected to an absorption column by a settler.

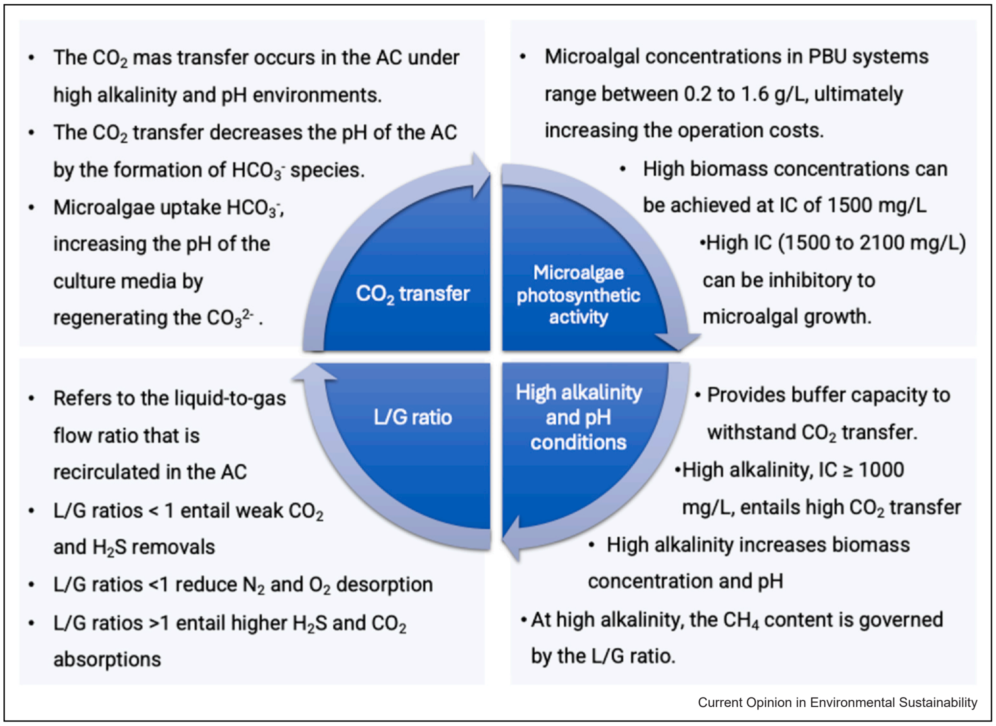
CO_2 contents as low as 0.2%, successfully meeting the EN 16723:2018 thresholds.

Indeed, microalgae cultivation requires a high water demand, increasing concerns for water security and agricultural competition, but the use of nutrient-rich effluents, namely centrate, the liquid fraction of digestates, could improve the system circularity [10]. Centrates have a high ammonia nitrogen content and require a posttreatment before being discharged [20]. Moreover, their high buffer capacity and low heavy metal concentration can trigger their use for PBU [10,21]. Hence, an integrated PBU process results in a dual environmental benefit by recycling nutrients to produce biomass and biomethane.

To date, centrates derived from the AD of food waste, pig farms, and sewage treatment plants have been used during PBU. Even if the nutrient removal efficiency depends on the centrate nutrient load and microalgae species, total nitrogen and phosphorus removals ranging between 70–100%, and 50–100% have been successfully achieved, respectively [22–25]. This intense nutrient uptake has resulted in microalgae biomass productivities of 0.2 g/Ld or 22 g/m^{2}d, which still remain low for commercial exploitation [22].

The use of centrates during PBU has been limited to robust microalgae strains, usually tolerant to high ammonia concentration. Typically, microalgae species with poor commercial value dominate these cultivation broths, that is, *Chlorella* sp., *Chlorococcum* sp. *Desmodesmus* sp. *Scedenesmus* sp. and some cyanobacteria in a lower share, namely, *Anabaena* sp., *and Leptolyngbya* sp. [9]. This microalgae population dominance has directed biomass commercialization to biofertilizers or biostimulants [26–28]. However, wastewater/centrate cultivated microalgae fertilizers entail divergent opinions by the presence of heavy metals, antibiotics, and pathogens [29]. Nonetheless, García-Galán et al. [30], demonstrated that microalgae biomass cultivated in wastewater fulfilled the thresholds set by the European legislation for fertilizers and did not entail a potential risk for the cultivated crops. Similarly, Álvarez-Gónzález et al. [31], demonstrated that microalgae biomass cultivated in wastewater was successfully used as fertilizer for lettuce crops. Both microalgae biomass and the cultivated crops fulfilled the thresholds set by the European legislation for fertilizers during pathogens and contaminants of emerging concern analysis. Interestingly, Cd was detected in all the cultivated crops, suggesting Cd was present in the freshwater used for watering rather than in the microalgae biomass. Nevertheless, the use of centrates derived from distillery and food waste represent a less controversial nutrient source as a result of their low concentration of heavy metals and chemicals [27,32].

Figure 3



Key operational parameters limiting the process of PBU.

Process limitations and opportunities

PBU still faces some operational drawbacks and limitations for further implementation on a medium and large scale. Indeed, the composition of the upgraded biomethane depends on the mass transfer of CO_2 to the culture broth, which is related to operational parameters such as alkalinity, pH, liquid-to-gas (L/G) ratio within the column, and on the photosynthetic activity of microalgae (Figure 3).

The diurnal variability of microalgae metabolism, as well as the seasonal change across the year, significantly impact O_2 content in biomethane and microalgae growth. Different strategies have been implemented to decrease O_2 in the upgraded biomethane namely modifying L/G ratio in the absorption column, enhancing mixing in the reactor to foster O_2 stripping, the addition of a O_2 stripping column, and operating under mixotrophic cultivation [33]. However, in most cases, the CO_2 content in the upgraded biomethane is increased, and higher alkalinity and an improved microalgae metabolism would be necessary to effectively remove CO_2. Moreover, during winter, microalgae growth is reduced, compromising the overall system operation. Even if the use of a greenhouse has been proposed to increase the temperature during cold winters [34], light availability and the high investment costs of greenhouses could limit both microalgae growth and the economic feasibility of PBU.

Additionally, microalgae cultivation requires high superficial areas for HRAP deployment. For instance, a biogas plant producing 300 Nm^3_{biogas}/h would require an illuminated area of 13.4 ha; hence, 1 Nm^3_{biogas}/h requires 621 m^2 [17]. Inherently, larger biogas plants would require HRAPs with higher superficial area, increasing concerns with food competition. Even if installing the HRAP inside the biogas plant could potentially reduce its footprint, PBU would not be appropriate in biogas plants with limited land space.

Indeed, like most upgrading technologies, PBU still has drawbacks and limitations, but an integrated approach for centrate treatment, CO_2 biofixation, and high-value compounds production can overcome the above-mentioned limitations. Thereby, strategies to enhance CO_2 mass-transfer to the liquor broth and microalgae metabolism to efficiently uptake CO_2, are mandatory to reduce the size of the plants, boosting the feasibility of PBU and accelerating commercialization.

Position in the market

Today, PBU has a TRL of 6–7 and has been successfully demonstrated at semi-industrial scale [16]. It has a low-energy demand, and its operating and maintenance costs are estimated < €0.1/Nm^3 of the upgraded biomethane. The investment cost of a standard PBU plant with a biogas production capacity of 300 Nm^3/h can reach up to

€6000/Nm^3, including the concentration and drying of the microalgal biomass. The commercialization of microalgae biomass for fertilizer production can generate revenues within five years, but the production of high-value compounds, namely pigments or nutraceuticals, could decrease the time to revenues.

In addition, meeting the ambitious biomethane production of 35 bcm in 2030 will result in 46 Mt of biogenic CO_2 that could be potentially exploited. Hence, PBU could directly use the biogenic CO_2 to produce high-value chemicals without requiring any additional separation or compression step for storage and transportation [35].

To date, a demo-scale PBU biorefinery composed of a 300 m^2 HRAP and a 0.5 m^3 column is operating with a capacity to treat 13 m^3_{biogas}/d entailing high biomass productivity and complete H_2S removal [16]. However, process scaling is needed to consolidate the feasibility of this technology.

Improving photosynthetic metabolism via nanoparticle addition

The addition of nanoparticles (NPs) to improve microalgae biotechnology has gained particular attention to boost microalgae growth and lipid accumulation [36]. Specifically, NPs synthesized via hydrothermal carbonization (HTC) of olive-mill wastewater (OMWW) have demonstrated great biocompatibility and outstanding biostimulant effect during microalgae growth.

HTC process is a well-known technology to produce a solid fertilizer, better known as hydrochar, and a nutrient-rich liquid, better known as tar, from sludge [37]. While tar has demonstrated to improve biogas yield during AD, both hydrochar and tar have been used to improve plant growth mainly due to the presence of essential micronutrients and bioactive compounds [38,39].

The addition of carbon-coated iron NPs synthesized via HTC has demonstrated an outstanding performance during PBU, increasing biomass productivities by two-fold while simultaneously fostering CO_2 removal efficiencies. This boosted effect has been identified at doses of 70 mg/L in *Chlorella sorokiniana* and mixed microalgae-bacteria batch cultures [40]. The scale-up of the process in a 180-L pilot-laboratory-scale system supplemented with 70 mg/L NPs entailed a two-fold increase in biomass concentration (> 3.5 g/L) and an increased CO_2 removal efficiency from 86% to 92%. Increasing NP dosage to 140 mg/L resulted in an improved biomass productivity up to 48 g/m^2d with CO_2 removal efficiencies > 98% [41].

The effect of tar was also tested during PBU, and the addition of 1 $mL_{tar}/L_{centratefed}$ presented similar stimulant effects on microalgae growth and CO_2 removals as the NPs. The long-term effect of tar at daily supplementation of 2 $mL_{tar}/L_{centratefed}$ supported a biomass productivity of 90 g/m^2d and CO_2 removal efficiencies > 95%, despite harsh operational changes within the system [42]. Interestingly, the suppressed supplementation of tar reduced the CO_2 removal efficiency to 82% and the biomass concentration to 1.6 g/L underscoring the potential of tar to maintain a stable PBU operation.

The use of NPs could represent a potential risk for biodiversity and compromise the environment integrity. Toxic effects have been observed with TiO_2, CuO, ZnO, Cr_2O, AgNPs, CoNPs, and some iron oxide NPs [43–45]. Indeed, NP toxicity depends on their crystal phase and surface physicochemical properties, but their biocompatibility can be increased by adding a carbon layer of OMWW [46]. The size of OMWW-covered NPs typically ranges between 70 and 100 nm, preventing their internalization to microalgae cells, thereby the NPs can be recovered during the harvesting of biomass. However, if high-value compounds production from microalgae is intended, extraction methods are highly selective, that is, pigments, and the NPs would remain in the residual biomass. Hence, either the produced microalgae biomass or the residual biomass containing the NPs could be further used as plant fertilizers or AD boosters [37]. Nonetheless, the high stability of NPs would result in an expanded lifespan, and further studies regarding the final fate of NPs are necessary before scaling this technology.

Conversely, NP synthesis via HTC is a high-energy-demand process. Thus, the valorization of the produced syngas during HTC and the production of biofuels and/or high-value compounds, along with the production of the microalgal stimulating tar, are necessary to counteract the production costs of the overall PBU process [38].

Photosynthetic biogas upgrading under a biorefinery concept

The addition of NPs to microalgae cultures has diversified the valorization of biomass to the production of biofuels, nutraceuticals, pigments, and environmental applications, including efficient nutrient uptake and CO_2 capture [47]. Nevertheless, pigment production stands as a more cost-effective and attractive pathway to improve PBU profitability as the market value of microalgae-based pigments is projected to grow to €7.2 billion by 2034 [48].

Typically, pigment-producing microalgae have slow growth rates and low biomass productivities, resulting in high production costs. Hence, NP supplementation stands as a promising technique to improve their growth and pigment yield. Recently, Au, OMWW-based, MoS_2, and ZnO NPs have improved the pigment content of *Chlorella zofingiensis*, *Coelastrella thermophila* var. *gobulina*, *Dunaliella salina*, and *Haematococcus lacustris* [48–50].

Despite the limited literature regarding integrated PBU and pigment production, the supplementation of 1 mL/L of tar derived from OMWW to *Coelastrella thermophila* var. *gobulina* cultures devoted to PBU increased the total carotenoid (TC) content to 838 μg/g [48]. Increasing tar concentration to 3 mL/L mediated a TC content of 1116 μg/g while maintaining a biomass productivity of 1.2 g/Ld. This confirmed the effect of tar on stimulating growth rate, which was supported by the accelerated nutrient consumption and a drastic decline in the photosynthetic activity, leading to an early endogenous phase where *C. thermophila* can synthesize carotenoids [48].

Certainly, the validation of these batch results in a continuous system using real centrate as culture media is needed to assess the feasibility of the integration of pigment production during PBU.

Conclusions

PBU has been successfully demonstrated at a semi-industrial scale. However, there are still some constrains to address, namely process stability and microalgal metabolism. Recently, nanoparticle supplementation has emerged as a cost-effective technique to stimulate microalgae metabolism, triggering stable system operation and resulting in a high-quality biomethane that meets the European standard for grid injection. In addition, the improved biomass production and valuable chemical products, that is, pigments, mediated by nanoparticle supplementation, could accelerate PBU commercialization. Nonetheless, the potential risks of nanoparticle supplementation needs to be studied before reaching industrial commercialization.

Data Availability

No data were used for the research described in the article.

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.

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- •• of outstanding interest

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