

Chemo-bio catalytic strategy for upcycling carbonaceous waste to value-added products

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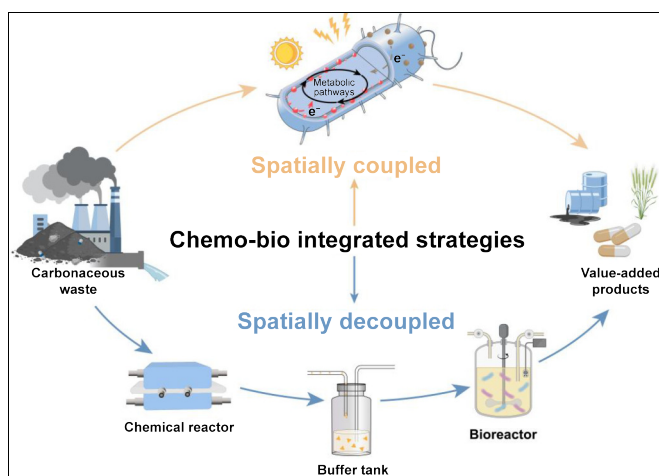


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ABSTRACT: The conversion of carbonaceous waste into value-added chemicals represents a sustainable waste management paradigm with the potential to alleviate mounting global carbon emission pressures and environmental crises. The inherent chemical inertness of carbonaceous wastes, such as CO₂ and plastics, necessitates harsh recycling conditions, making it challenging to achieve both high conversion and high selectivity, which severely limits the process efficiency. Conversely, enzyme-driven biotransformations operate under ambient conditions with exceptional selectivity, but are hindered by narrow substrate scope and slow kinetics. By synergistically integrating the high efficiency of chemical catalysis with the precision of biosynthetic pathways, chemo-bio catalytic integrated strategies offer a promising alternative. These approaches enable continuous energy input for enzymatic upgrading of carbon waste through chemical energy-transduction mechanisms, while converting complex carbonaceous materials into small molecule intermediates that are readily assimilated by microbes, thus overcoming limitations in both substrate scope and energy supply. This perspective systematically summarizes recent advances and evolving trends in chemo-bio integrated strategies for the valorization of carbonaceous waste. We provide a comprehensive analysis from the perspectives of spatially coupled and spatially decoupled systems, and further examine persistent challenges related to efficiency enhancement, system robustness, energy utilization, and scalable integration.



KEYWORDS: chemo-bio catalytic strategy, carbonaceous waste, value-added products, microbial metabolism

1 Introduction

As global carbon emission pressures continue to mount, the accumulation of carbonaceous waste has posed unprecedented challenges to environmental safety and resource sustainability^{1–3}. From the vantage point of a circular economy, these conventionally defined environmental pollutants should instead be re-conceptualized as repositories of significant valorization potential^{4–6}. However, conventional thermochemical conversion of

carbonaceous waste streams (e.g., CO₂ and plastics) is generally confronted with high activation barriers and poor reaction selectivity, which severely constrains the efficiency of their conversion into value-added products^{7–9}. To realize the targeted upcycling of carbonaceous waste into value-added chemicals, the development of catalytic strategies that combine exceptional activity with exquisite selectivity has thus become a central imperative in contemporary green chemistry and sustainable technology^{10, 11}. Addressing this challenge not only advances fundamental science but also provides critical technological scaffolding for the attainment of carbon-peak and carbon-neutrality commitments.

In recent years, emergent chemical conversion strategies such as electrocatalysis and photocatalysis have demonstrated considerable promise for the valorization of carbonaceous waste^{12–14}. With their efficient reaction kinetics, these chemical methods enable rapid activation of inert carbonaceous waste^{15–17}. These purely chemical approaches, however, are constrained by harsh operating

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conditions, limited energy efficiencies, and an inadequate capacity for fine-tuning product selectivity. Conversely, the enzyme-based biotransformation operates under benign ambient conditions and offers exquisite stereoselectivity, yet remains hampered by sluggish reaction rates and restricted substrate scopes¹⁸. These complementary limitations have galvanized efforts to devise hybrid chemo-bio or biomimetic catalytic systems that synergistically integrate the efficiency of chemical activation with the precision of biosynthesis^{19–21}. By orchestrating intimate couplings between tunable chemical steps and microbial metabolism, researchers can program energy and materials relay cascades that continuously feed redox equivalents into living catalysts while converting structurally complex wastes into bio-assimilable intermediates. This strategic merger not only alleviates the energetic and substrate bottlenecks inherent in each individual platform but also exploits the tunable reactivity of chemical modules to channel flux toward high-value metabolites with minimal by-product formation^{22–24}. The resultant chemo-bio integrated systems thus establish a rational blueprint for the green, efficient, and selective upcycling of carbonaceous waste into value-added chemicals.

The chemo-bio integrated strategy offers a transformative paradigm for the value-added valorization of carbonaceous waste by strategically fusing the high throughput of chemical catalysis with the precision of biosynthesis. At its core, on the one hand, this strategy exploits the tunable reactivity of chemical modules to construct a rationally designed electron-transport and energy-relay pathway^{25–31}; on the other hand, the reducing equivalents and active intermediates generated by the chemical modules are allowed to be seamlessly funneled into downstream biosynthetic pathway^{32–38}. This synergistic effect achieves three breakthroughs: First, the chemical pretreatment depolymerizes complex carbonaceous waste into readily assimilable small-molecule precursors, thereby extending the substrate envelope far beyond the native scope of microbial enzymes. Second, redox equivalents and adenosine triphosphate (ATP) equivalents generated *in situ* by the

chemical modules provide a continuous energy input that sustains microbial metabolism. Most importantly, the unique enzyme cascade reactions in the biological system impose exquisite control over product selectivity, transforming the unavoidable side reactions that plague traditional chemical routes into kinetically favored pathways for the targeted synthesis of desired products. The resulting “chemical disassembly–biological reconstruction” cascade not only transcends the intrinsic kinetic vs. selectivity trade-offs of individual strategies but also opens up a new paradigm for resource-efficient and product-selective upcycling of carbonaceous waste.

This perspective systematically reviews the latest research trends and development trajectory of chemo-bio integrated strategies for the value-added valorization of carbonaceous waste. By dissecting representative case studies across multiple dimensions, we illuminate both cutting-edge advances and the key scientific challenges and technological route for future development. The perspective is structured around two complementary design: spatially coupled and spatially decoupled integrated strategies (Fig. 1). For spatially coupled integrated systems, we focus on the mechanisms of direct energy and electron transfer at the chemical–microbial interface, systematically evaluating the architectural principles and performance metrics of representative platforms (e.g., photocatalysis-microbe hybrid and electrocatalysis-microbe hybrid systems). We further chart a forward-looking on future directions centered on interfacial engineering and the maximization of energy-transfer efficiency. In the realm of spatially decoupled integrated strategies, we highlight the unique advantages of modular design in circumventing thermodynamic and kinetic incompatibilities between chemical and biological steps and highlight the latest research advances in representative decoupled systems (e.g., electrocatalysis–fermentation coupling and chemical catalysis-bioconversion tandem systems). Concurrently, from an engineering vantage point, we provide a prospective analysis of the scale-up challenges and intensification opportunities that will dictate the industrial realization of decoupled systems.

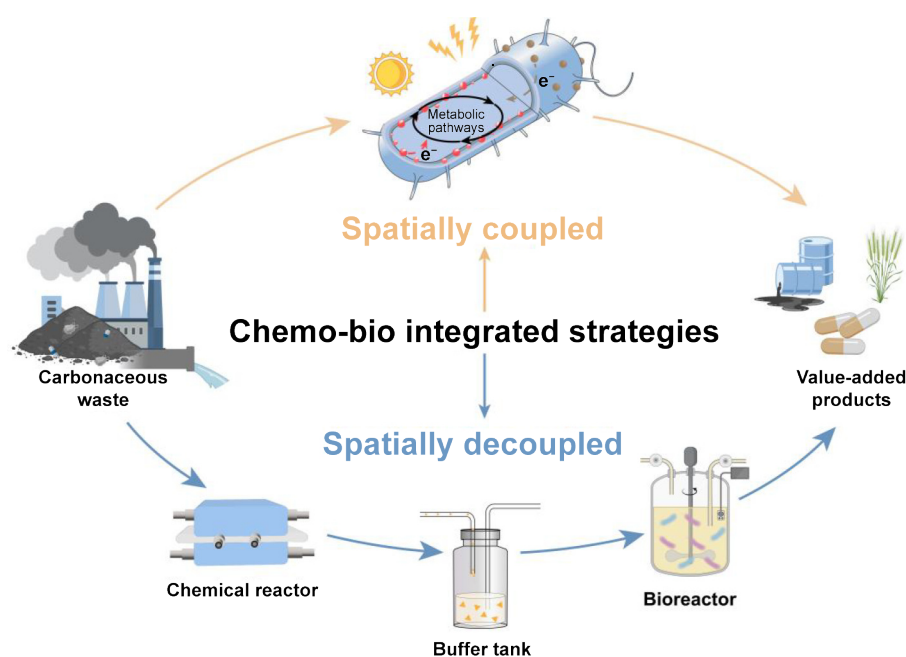


Fig. 1. Schematic illustration of chemo-bio catalytic strategy for upcycling carbonaceous waste to value-added products.

2 Spatially coupled chemo-bio integrated strategies

A spatially coupled chemo-bio integrated strategy refers to a design paradigm in which the chemical catalytic module and the microbial metabolic module are co-localized within a shared physical space. Central to this strategy is the construction of an efficient chemo-bio interface that enables renewable energy inputs (e.g., light and electricity) captured and transduced by the chemical module to be delivered directly to the microbial cells, thereby driving the upcycling of carbonaceous waste into high-value products. Ideally, such an integrated configuration enables *in situ* energy and mass transfer with minimal dissipation, maximizing overall conversion efficiency.

Initially, Sakimoto et al.³⁹ pioneered the chemo-bio integrated strategy by constructing a CdS-*Moorella* (*M.*) *thermoacetica* hybrid that converts typical gaseous carbonaceous waste CO₂ into the value-added chemical acetic acid (Fig. 2a). Upon visible-light irradiation, the photocatalyst CdS generates photoelectrons which are directly harvested by *M. thermoacetica* to power reductive acetogenesis, achieving a yield up to ~90% (Fig. 2b). Inspired by this work, Pi et al.²⁷ engineered the halotolerant *Vibrio* (*V.*) *natriegens* to heterologously biomineralize CdS *in situ*, thereby constructing a CdS-*V. natriegens* hybrid. By further introducing a heterologous 2,3-butanediol (BDO) biosynthetic pathway, they achieved visible-light-driven value-added valorization of organic wastes in sewage to the high-value product BDO, underscoring the versatility of the hybrid strategy.

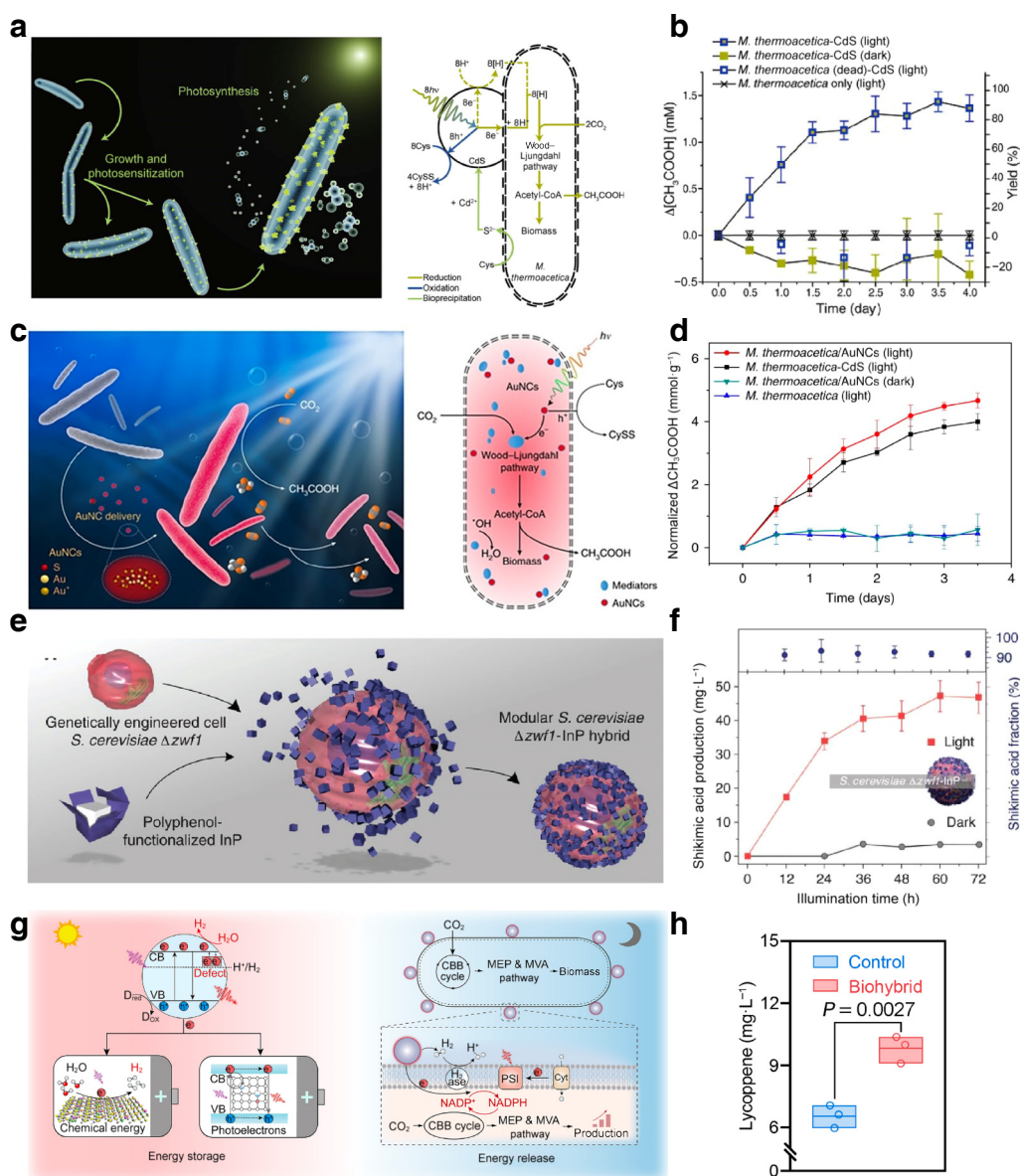


Fig. 2. Spatially coupled chemo-bio integrated strategies. **a**, Schematic of the CdS-*M. thermoacetica* reaction system. **b**, Photosynthetic production of acetic acid by CdS-*M. thermoacetica* hybrids and deletional controls. Reproduced with permission from Ref.³⁹, © American Association for the Advancement of Science 2016. **c**, Schematic diagram of the AuNC-*M. thermoacetica* hybrid system. **d**, Normalized photosynthetic production of acetic acid by AuNCs-*M. thermoacetica* under continuous low-intensity illumination and in dark conditions. Reproduced with permission from Ref.⁴⁰, © Zhang, H. et al. 2018. **e**, Schematic of the fabrication of InP-*S. cerevisiae*. **f**, Shikimic acid production under light and dark conditions. Reproduced with permission from Ref.⁴¹, © Guo, J. L. et al. 2018. **g**, Schematic illustration of the spatiotemporally solar-decoupled biohybrid. **h**, Yield of lycopene biosynthesis. Reproduced with permission from Ref.⁴³, © Elsevier Inc. 2024.

Researchers have since diversified photocatalytic materials and improved the electron transfer at chemo-bio interfaces to enhance electron utilization efficiency and robustness of the hybrid. For example, Zhang et al.⁴⁰ constructed an Au nanocluster (AuNC)-*M. thermoacetica* integrated system in which the photocatalyst was internalized into the cytoplasm of *M. thermoacetica*, bypassing transmembrane electron transfer limitations and achieving efficient valorization of CO₂ under visible light (Figs. 2c and 2d), with a CO₂-to-acetate yield of approximately 4.4 mmol·g⁻¹ after 4 days of irradiation. Guo et al.⁴¹ reported a functionalized InP-*Saccharomyces (S.) cerevisiae* integrated system that exploits surface-adhesive and conductive polyphenols to anchor the photocatalyst intimately to the cell envelope while constructing an low-resistance electron conduit, thereby enabling light-driven synthesis of the high-value product shikimic acid (Figs. 2e and 2f), achieving a conversion yield of approximately 90%. Zhang et al.⁴² further developed an organic semiconductor-*Ralstonia (R.) eutropha* integrated system, in which the organic semiconductor exhibits strong quadrupolar interactions with cell membranes and effectively promotes charge separation as well as transmembrane injection of photogenerated electrons, culminating in the high-yield conversion of CO₂ to poly(β-hydroxybutyrate) bioplastics.

Although photocatalysis-based chemo-bio integrated strategies have achieved remarkable success in the value-added valorization of carbonaceous waste, they remains constrained by environmental factors such as diurnal light fluctuations and the day-night cycle. To address this bottleneck, we constructed a sunlight decoupled hybrid system⁴³. By integrating a persistent luminescent photocatalyst with photoautotrophic microorganisms, we enabled all-weather synthesis of the high-value product lycopene from CO₂ waste, thereby providing a new avenue for extending photocatalytic reactions beyond *in situ* illumination (Figs. 2g and 2h) and achieved a lycopene yield of 10–11 mg·L⁻¹.

Electrocatalysis-based chemo-bio integrated strategies likewise provide an effective route for the value-added valorization of carbonaceous waste. Liu et al.⁴⁴ reported an electro-microbe system that drove an artificial carbon-fixation process, enabling the value-added valorization of CO₂ into poly(3-hydroxybutyrate) (PHB) as biomass and value-added liquid fuels, including isopropanol, isobutanol, and 3-methyl-1-butanol. In this system, a cobalt phosphate anode and a NiMoZn alloy cathode were employed to efficiently drive the oxygen evolution reaction and the hydrogen evolution reaction, respectively. The generated H₂ was further captured by engineered *R. eutropha*, which converted CO₂ into biomass and fuels at a low ambient concentration. Li et al.²⁵ bypassed heterogeneous electrocatalysis entirely by interfacing an electrode with engineered *Shewanella oneidensis*, achieving the valorization of CO₂ to the complex C₄ compound malic acid.

Additionally, photoelectrochemical systems that integrate the high efficiency of photocatalysis with the precise control of electrocatalysis have emerged as promising platforms for carbonaceous waste upcycling. For example, Kim et al.²⁶ integrated the photoelectrochemical conversion of CO₂ with enzymatic plastic waste valorization in a single solar-driven system, which offers a new avenue for the mitigation of two carbonaceous waste streams (CO₂ and plastics) to produce useful products in the form of fuels and chemicals.

Despite the unique advantages of spatially coupled chemo-bio integrated strategies in direct energy transduction and the breakthroughs achieved in the value-added valorization of various

carbonaceous wastes, several challenges remain for practical application. (1) The disparate optimal operating windows of chemical reactions and microbial metabolism (e.g., light intensity, current density, pH, and temperature) complicate the co-optimization of the system, thereby limiting the conversion efficiency and stability. (2) Photocatalytic systems are susceptible to environmental perturbations such as diurnal cycles and climatic variability, resulting in intermittent electron supply, whereas electrocatalysis suffers from reduced microbial activity at high current densities as well as electrode fouling or passivation. (3) Accumulation of inhibitory by-products within the shared reaction space. (4) Insufficient transmembrane electron transfer efficiency across the chemo-bio interface. (5) Progressive loss of material stability and cellular adaptability over extended operation. All of these factors collectively constitute major bottlenecks for industrial scale-up and widespread deployment.

Future efforts should advance along three complementary trajectories: (1) Interface engineering optimization: The molecular level surface functionalization, nanostructure regulation, and biomimetic materials are suggested to be applied to enhance the transmembrane electron-transfer efficiency and reduce interfacial charge losses. (2) Integration of photoelectric modules with energy storage units: Hybrid systems that integrate energy storage components will be developed, and these systems will synchronize photoelectric conversion and storage to guarantee stable productivity under diurnal or intermittent operation. (3) Dynamic regulation of the reaction microenvironment: Adaptive materials or microfluidics technologies should be utilized to adjust local pH, ionic strength, and substrate concentrations in real time, thereby buffering the disparities between chemical and biological process.

3 Spatially decoupled chemo-bio integrated strategies

In contrast to spatially coupled strategies, the spatially decoupled chemo-bio integrated strategy adopts a modular design philosophy, constructing discrete chemical and biotransformation units that operate in tandem to form a stepwise cascade. The strategy proceeds in two stages: First, a dedicated chemical catalytic module converts recalcitrant carbonaceous waste streams (e.g., plastics and CO₂) into microbially accessible small molecule intermediates, including organic acids and syngas. Second, these intermediates are precisely delivered to a separate biosynthetic module, wherein microbial cell factories execute the targeted synthesis of high-value products. This decoupled design not only effectively overcomes constraints arising from incompatible reaction conditions, but also markedly broadens the substrate scope and product diversity through modular recombination, thereby offering an innovative solution for building flexible and efficient biomanufacturing platforms based on the carbonaceous waste valorization.

As a representative example, Zheng et al.⁴⁵ proposed a modular electrocatalysis-fermentation integrated system in which CO₂ electrolysis and yeast cultivation are physically segregated, enabling the high-value valorization of CO₂ waste into glucose or fatty acids (Figs. 3a and 3b). The designed solid electrolyte reactor rapidly converts CO₂ into pure acetic acid intermediates at an industrially relevant current density, which is then fed as the sole carbon source to engineered *S. cerevisiae*. This spatial separation precludes the product overaccumulation, thereby averting metabolite induced

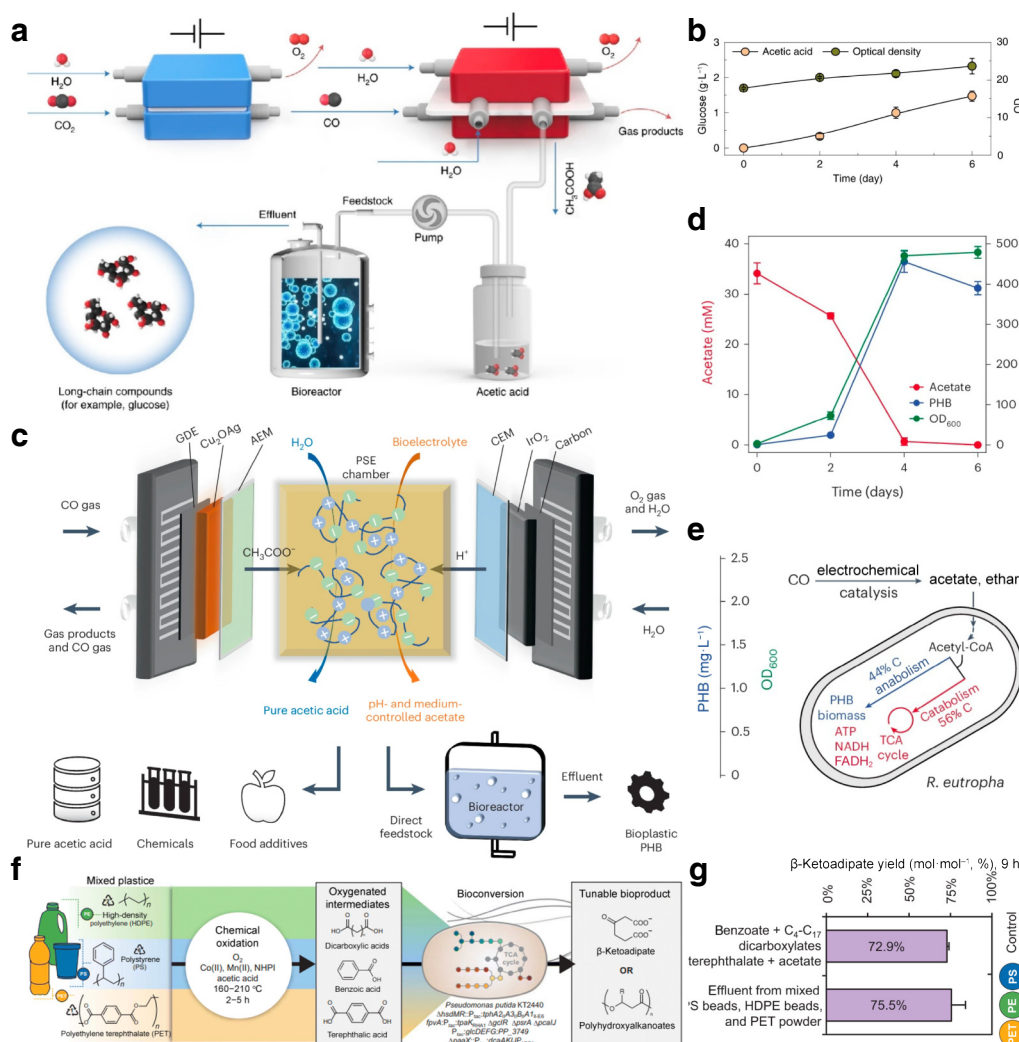


Fig. 3. Spatially decoupled chemo-bio integrated strategies. **a**, Schematic of the *in vitro* artificial sugar synthesis system. **b**, Electro-generated pure acetic acid as the direct feedstock for engineered strain *S. cerevisiae* LY031 to produce glucose (OD = optical density). Reproduced with permission from Ref.⁴⁵, © Zheng, T. T. et al. 2022. **c**, Schematic of a porous solid electrolyte (PSE) electrolyser with diverse product chains from electrosynthesized acetate. **d**, The growth of *R. eutropha* on acetate and the generation of PHB by a representative cultivation batch. **e**, Conversion of CO reduction products to multicarbon products in biocatalyst (*R. eutropha*). Reproduced with permission from Ref.⁴⁶, © Wi, T. U. et al. 2024. **f**, Schematic of the upcycling of mixed plastic waste through tandem chemical oxidation and bioconversion. **g**, Utilization of intermediates in effluent from mixed polystyrene (PS) beads, high-density polyethylene (HDPE) beads, and PET powder by AW307 and β -ketoadipate yields from the same or control mixtures. Reproduced with permission from Ref.⁴⁷, © Sullivan, K. P. et al. 2022.

microbial inhibition and enabling the efficient biosynthesis of glucose or fatty acids. Wi et al.⁴⁶ proposed a chemo-bio integrated system based on a porous solid electrolyte reactor, achieving the value-added valorization of CO to C_{4+} poly(β -hydroxybutyrate) bioplastics. In this system, the porous structure of the solid electrolyte allows the *in situ* dissolution of electro-generated acetate into a purpose-designed biocompatible buffer, obviating complex downstream processing. The resulting acetate solution is directly supplied to a downstream bioreactor, driving *R. eutropha* to synthesize the high-value product bioplastics PHB (Figs. 3d and 3e) and achieved a PHB titer of $\sim 510 \text{ mg L}^{-1}$.

The spatially decoupled strategy is not confined to electrocatalytic transformations; it readily extends to chemocatalysis, hydrolysis, and other reaction modalities. For example, Sullivan et al.⁴⁷ proposed a chemocatalysis–biocatalysis integrated strategy for the value-added valorization of mixed plastic wastes (Figs. 3f and 3g). In this system, unsorted mixed plastics undergo metal-catalyzed autoxidative depolymerization to yield a

suite of oxygenated small-molecule intermediates such as benzoic acid, terephthalic acid, and various aliphatic diacids, which are subsequently funneled by engineered *Pseudomonas putida* into a single chemical product with a yield of $\sim 75\%$, thereby providing a new route for converting mixed heterogeneous plastic wastes into useful chemicals. Witt et al.³² employed chemical hydrolysis to efficiently cleave polyamide plastics into monomers and oligomers, which were subsequently funneled through engineered *P. putida*, achieving the value-added conversion from waste plastics to poly(β -hydroxybutyrate). These diversified chemical approaches expand the versatility of spatially decoupled chemo-bio integrated strategies to the value-added valorization of a broad spectrum of carbonaceous waste.

Notably, Zhao et al.³³ proposed a transformative valorization workflow for carbonaceous waste that leverages the spatially decoupled strategy to chain discrete reaction modules. By integrating the mechanical milling, chemical conversion, and biosynthesis into a single unified workflow, their process enabled

efficient conversion of complex municipal sewage sludge into high-value products. This work underscores the distinctive potential of spatially decoupled chemo-bio integrated strategies to orchestrate heterogeneous reaction types and construct complex transformation systems.

By virtue of the modular separation, the spatially decoupled chemo-bio integrated strategy effectively eliminates thermodynamic mismatches between chemical catalysis and microbial metabolism with respect to temperature, pH, ionic strength, and reaction medium, while markedly broadening the spectrum of convertible substrate and attainable product. In current studies, the chemical module typically furnishes high purity intermediates that can be directly assimilated into the biological module, thereby streamlining the process and improving overall energy efficiency. However, this strategy still faces several practical challenges: (1) the dynamic matching intermediate concentration and supply rate between the chemical and biological modules, (2) the long-term stability of interfacial reaction conditions and further improvements in the energy efficiency of the chemical module, and (3) the integration and control of multiple modules under scaled-up operation.

Future research may proceed along the following directions: (1) Module operational matching: implementing precise process control to achieve real-time dynamic balance between intermediate generation and biological consumption, (2) Energy efficiency optimization: developing high efficiency and low energy cost chemocatalytic and electrocatalytic systems to reduce overall energy demand. (3) Reaction medium compatibility: engineering universal buffer systems suitable for both chemical conversion and biosynthesis to minimize conditioning adjustments. (4) System integration and scale-up: building modular production platforms capable of continuous operation, facile scale-up, and seamless integrating with upstream and downstream processes.

4 Conclusions

In summary, this perspective systematically reviews the key research advances in recent years on chemo-bio integrated strategies for value-added valorization of carbonaceous waste. We dissect the field along two principal axes: (1) Spatially coupled chemo-bio integrated, wherein chemical reactions and biological metabolism share a common physical space. Here we emphasize the underlying mechanisms of direct energy transfer and delineate performance optimization routes in both photocatalytic and electrocatalytic systems. (2) Spatially decoupled chemo-bio integrated. In this part, we highlights the elimination of condition conflicts and the expansion of substrate utilization through modular separation, and examines practical implementations and potential in electrocatalytic, chemocatalytic, and hydrolytic systems. Overall, by integrating the rapid kinetics of chemical processes with the high selectivity of biosynthesis, these two classes of strategies show marked advantages and broad application prospects in converting typical carbonaceous wastes such as CO₂ and plastics into high-value products.

Despite these advances, this field still faces numerous technical challenges on the path toward further development and industrial deployment. First, the compatibility of reaction conditions and overall system stability remain central hurdles: (1) In spatially coupled chemo-bio integrated systems, differences in the optimal operating conditions of chemical reactions and microbial metabolism limit overall efficiency and stability; by contrast, spatially decoupled chemo-bio integrated avoids direct conflicts in

reaction conditions yet requires long term dynamic matching between the chemical and biological modules in terms of intermediate concentration and actual reaction rate. (2) The chemical module is generally energy intensive, calling for high efficiency and low energy cost catalytic systems, while also enhancing mass transfer and anti-fouling capability under scale-up conditions. Moreover, the tolerance of materials and microorganisms under long term operation requires further optimization to cope with adverse effects arising from electrode passivation, by-product accumulation, and substrate impurities.

At the economic and industrial deployment levels, the process integration of chemo-bio integrated platforms with existing high emission industries (e.g., power plants, chemical plants, and plastic-recycling facilities) can reduce feedstock acquisition and separation costs while enabling synergistic carbon-reduction benefits. However, industrial off gases and effluents often contain inhibitory components such as SO₂, H₂S, and heavy-metal ions, necessitating the development of efficient and cost-effective pretreatment processes to ensure stable coupling between the chemical and biological modules.

Looking ahead, deep convergence of multidisciplinary knowledge is poised to unlock transformative potential in this field. Precision analytical chemical tools will enable online monitoring and dynamic optimization of reaction processes; advanced functional materials will markedly enhance interfacial electron transfer efficiency; next generation synthetic biology will expand the substrate utilization spectrum and stress tolerance of microorganisms; and adaptive artificial intelligence will deliver real time control and predictive fault management for complex integrated systems. Through the synergistic development of these technologies, chemo-bio integrated strategies are expected to achieve high efficiency, low energy cost, and sustainable industrial deployment across a broader range of carbonaceous waste scenarios, laying the foundation for a green, low-carbon circular economy.

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Data availability

Not applicable.

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Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

Use of AI statement

None.



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