PERSPECTIVE

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Wastewater Upgrading to Fuels: Routes and Challenges

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Abstract

This perspective outlines technical strategies for upgrading wastewater into fuels and value-added chemicals, emphasizing a shift toward circular economy and resource recovery through advanced processes like photocatalysis, electrolysis, and microbial technologies.

Keywords: renewable fuels, water-energy-carbon nexus, hydrogen, circular economy, water recovery.

In view of the constant increase of worldwide population, natural resources depletion, like water, has become an issue addressed from different corners, i.e. scientific, social, political and economic. In this context, water recovery and reuse are relevant not only to aid the achievement of sustainable development goal 6 but also poses the potential of aiding decarbonization and ensuring a circular economy. This perspective aims to summarize technical strategies to valorize wastewater through its upgrading towards fuels and value-added chemicals, e.g. hydrogen, methane, methanol, ethanol, formic acid, urea and ammonia.

Wastewater can be classified in three broad categories depending on source, composition (a) industrial wastewater (b) domestic wastewater/sewage and (c) stormwater runoff [1]. The information related to global wastewater generation is limited; however, it has been estimated 380 billion m^3 [2]. Wastewater composition depends on the source and goes from simple to very complex. Gray water from bathroom is an example of a very simple composition since it only contains soaps and microbes. Its complexity increases when mixed with kitchen wastewater because this adds lipids, proteins, minerals and salts [1]. Wastewater from industry is complex and it contains a wide variety of heavy metals, biological, organic and inorganic compounds as a function of the originating process, which can be from mining, automobiles, food, chemical, textiles, plastics, pharmaceutical or steel industry.

Let us take as an example of wastewater complexity that coming from the food industry, specifically from a chocolate manufacturing industry. Figure 1 shows a typical characterization. As observed, it contains over 1300 mg/L of total organic carbon (TOC), 95 mg/L of total nitrogen and of 36.82 mg/L of phosphates [3]. It is here, in its composition, where the potential for upgrading wastewater is enclosed.

For decades, wastewater recovery and reuse have focused only on the development of technologies to remove contaminants both, organic and inorganic, from water. This was always conducted searching for high mineralization rates and percentages, and

Submitted: 01 August 2025
Accepted: 11 August 2025
Published: 28 August 2025

Vol. 1, **No.** 1, 2025.
10.62762/JCERF.2025.342282

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Citation
Natividad, R. (2025). Wastewater Upgrading to Fuels: Routes and Challenges. *Journal of Chemical Engineering and Renewable Fuels*, 1(1), 3–8.

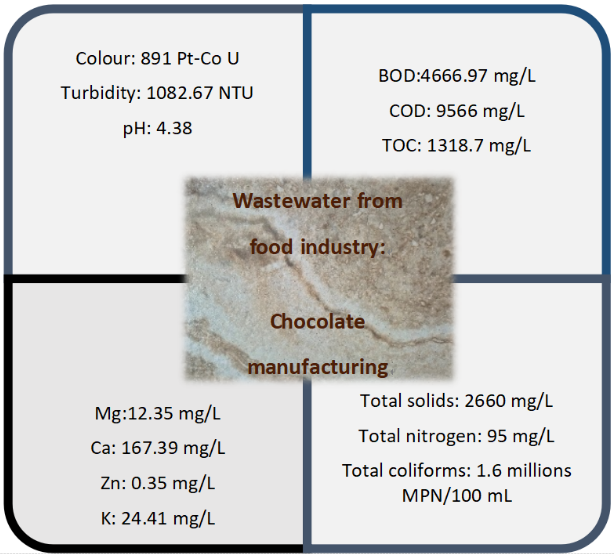

Figure 1. Typical composition of wastewater from a food industry: chocolate manufacturing.

elimination of the inorganic and biological load, to comply with environmental standards that in many countries overlook emerging contaminants or the disposal of toxic effluents for biomes.

It is approximately in the last decade that a change in paradigm began to emerge. In this new paradigm, to begin with, the term wastewater is substituted by residual water and is no longer considered a waste but a feedstock of novel processes aiming to re-use water or to produce alternative fuels (ethanol, methanol, formic acid) or precursors of value-added chemicals (oxalic acid, acetic acid, malonic acid, etc.). A typical train of processes to treat water consists of three stages, i.e. clarifying + biological treatment + advanced oxidation (when the composition includes recalcitrant pollutants). The biological treatment aims to remove pollutants and is conducted in bioreactors that use microorganisms to conduct aerobic or anaerobic digestion. However, there are some recalcitrant compounds that due to their chemical nature cannot be biologically degraded and their removal demands an advanced oxidation process.

Advanced oxidation processes (AOPs) are defined by the in-situ production of reactive oxygen species (ROS), particularly hydroxyl radicals, which are known to oxidize practically any organic compound. The advanced oxidation processes differ in the way the ROS are produced. In this sense, depending on the way the ROS are produced, an ACOP can be photochemical, electrochemical, chemical (Fenton, sulfates, H_2O_2, O_3), photo-electrochemical, and by ultrasound. Each of them pursuing the highest mineralization efficiency since it is the way to guarantee the lowest toxicity of the treated effluent.

The new vision of wastewater treatment, wastewater upgrading, pursues transforming wastewater into usable products, like substrate to produce biomass, fuels or added-value chemicals. This is an advanced concept in circular economy and resource recovery. Thus, from this perspective, the technologies to recover water must aim to conduct redox reactions in a selective way. This demands then the intervention of experts in materials science, chemical and environmental engineering, and renewable energies.

Figure 2 illustrates a proposed block diagram of a train of processes for wastewater upgrading. This includes inputs and outputs to the system. Each of these will be specific to the type of applied process and must be accounted for to establish the corresponding energy balance, environmental and economic impacts of the process. The first stage, pre-treatment, can be as simple as separating solids or more advanced if color and turbidity are to be removed. A unit operation to tune the wastewater composition by concentrating the fuel precursor compounds and removing the unwanted, can also be placed as pre-treatment, e.g. evaporation and/or membrane electrodialysis. Then becomes the stage for the upgrading that can be conducted by different means as will be below described. The stage of separation/purification consisting of one or several unit operations to recover the fuel or the generated value-added chemical. The emissions to air in each process are mainly due to energy consumption of fossil fuel type. For decarbonizing purposes, however, the use of solar light must be preferred. The emissions to the soil are for instance the separated solids, the heterogeneous catalyst or electrodes, even water dispose to the soil.

Although there might be other useful applications of treated wastewater, herein the perspective of wastewater transformation into fuels, fuels precursors and added-value chemicals will be summarized. In this context, the research trend is hydrogen production. Table 1 presents some examples found in the literature reporting the use of wastewater as feedstock to produce hydrogen and methane, as substrate for algas cultivation and to produce ammonia and formic acid. It is worth noting that algas can be used to produce hydrogen or oil to produce biodiesel and this is the reason for considering them in Table 1.

From the technologies presented in Table 1, the production of hydrogen by biological means, either

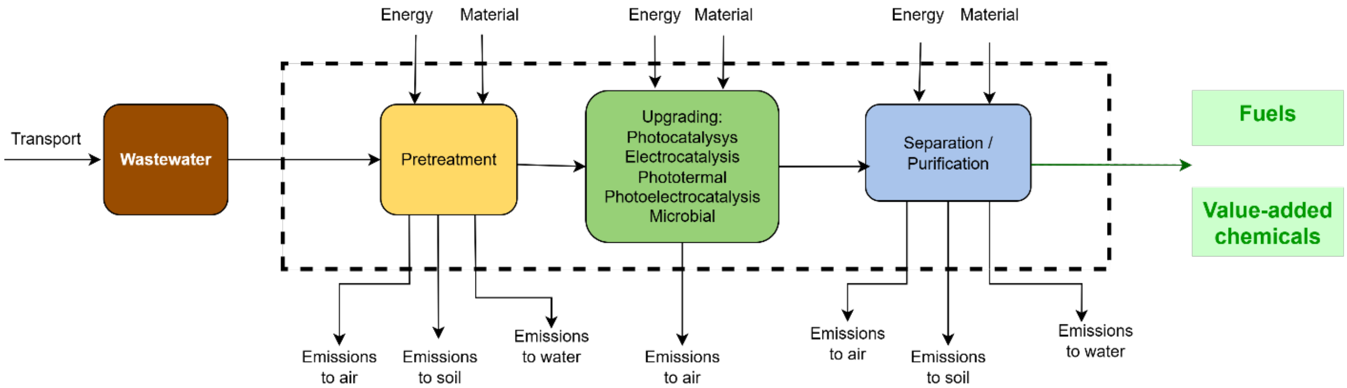

Figure 2. General block diagram for wastewater upgrading.

Table 1. Examples of wastewater upgrading towards fuels or their precursors.

Residual Water source	Fuel or precursor	Technology	References
Landfill leachate; Municipal sewage; Domestic Winery; Cellulose, glucose -based	H_2, CH_4	Microbial Electrolysis Cells; Biological	[4–8]
Sulphite wastewater (tannery, refinery); Brines	H_2	Water splitting, photocatalysis, photo-thermal, Photo-electrochemical conversion	[9–11, 23]
University, municipal and dairy	Biomass (algae)	Bubble columns to grow algas; Solar light; CO_2	[13]
Landfill leachate	Methane		
PET Bottles and simulated nitrate wastewater	Formic Acid	Paired electrolysis	[14]
PET Bottles and simulated nitrate wastewater	Ammonia	Paired Electrolysis	[14]

fermentation or Microbial Electrolysis Cell (MEC), is the most assessed and advanced technology for wastewater upgrading to a useful product: substrate to grow specific bacteria that produces hydrogen. The assessed wastewater as substrate in MEC is landfill leachate, municipal sewage, domestic water, winery residual water and cellulose and glucose-based water [4–7]. This is a versatile technology, the efficiency will be affected by the wastewater source, i.e. composition, microorganism and the electrodes material [5]. The electrodes material is of paramount importance, not only for the efficiency of the process but also for its cost.

Regarding the biological processes, i.e. fermentation and anaerobic digestion, to produce hydrogen and methane from wastewater, they benefit from wastewater rich in carbohydrates, lipids and proteins [8]. Thus, an effluent from food industry or landfill leachates represents a viable feedstock for this process. The base of this process is the bacteria to produce the gas fuel. Thus, in this case, wastewater is upgraded to substrate to grow specific bacteria, which can be cultivated in darkness or under light. This will be determined by the type of bacteria. This is an important remark because the input of energy in any way to produce fuels must not be overlooked because it affects not only the energy balance of the process but also its environmental impact categories.

Water splitting is another reported process for hydrogen production and the literature in this regard is vast, either by electrochemical or photocatalytic means. This is not the case, however, when wastewater instead of synthetic, deionized or distilled water is used. In this sense, [9] and [11] used sulphite wastewater to produce hydrogen via water splitting and photocatalysis, respectively. For the same process conducted via photocatalysis or electrochemistry, [10] stated the feasibility of using brine waters (from ocean, desalination industry and brackish groundwater). The hydrogen production rate in the first case was 17 mL/h. Municipal wastewater, previously treated in a biological reactor, was also demonstrated to be upgraded to O_2 and H_2 by electrolysis [12]. This work is a good example of recovering water and using it for different purposes.

Generally speaking, an advantage of both photocatalytic and electrochemical processes is the vast existing literature regarding catalysts, electrodes, electrochemical reactors and photo-reactors. An alleged disadvantage, however, is the typical slow kinetics associated with photocatalysis. This, however, might represent the main advantage for producing value-added chemicals, like formic acid, acetic acid and oxalic acid. At this moment, photocatalysis alone is not a viable process to commercially produce hydrogen because the efficiency is below 5-10%, which has been reported as an indicator of viability [15, 16]. This is the reason for being typically combined with thermal activation.

Photoelectrochemical systems merge the positive characteristics of both processes, photocatalysis and electrochemistry. These systems have been extensively studied and allow the production of hydrogen, carbon monoxide, methane, methanol, ethylene, ethanol and ammonia, by water splitting, carbon dioxide reduction and nitrites reduction at the cathode and the upgrading of organic compounds like urea, glycerol and phenols by oxidation at the anode [17, 18]. Once again, the research of these systems using wastewater as feedstock is scarce and the closest approach that might also be feasible is the analysis of water characterized by one compound [18]. Under such an analysis, for example, urea can be oxidized to hydrogen at the anode, while H_2 can also be produced by CO_2 reduction at the cathode [19]. In the context of hydrogen production, urea oxidation reaction is an efficient alternative to water splitting [20]. This approach is known as paired photo-electrolysis or paired electrolysis and another important example is the reduction of nitrate to ammonia in the cathodic chamber concomitantly with the oxidation of polyethylene-terephthalate-derived ethylene glycol (PET) towards formic acid at the anodic chamber [14]. This paired process led to excellent yields, 83% for formic acid and 80% for ammonia. For these results to be achieved, however, PET bottles were hydrolyzed with KOH prior anodic oxidation. Then a separation/purification stage, like evaporation, is added to obtain formate [14].

Worldwide, ammonia is extensively produced and contributes around 1% of total greenhouse gas emissions (GHG) [21]. This urges the development of green processes for its production. In the context of renewable fuels, ammonia is considered an important hydrogen carrier because it poses a hydrogen density (ca. 121kg H_2/m^3) higher than liquid hydrogen (ca. 71kg H_2/m^3) and gas hydrogen (ca. 40kg H_2/m^3) [21]. Ammonia can also be used directly as fuel or blended with others like methane [22]. The synthesis of green ammonia has also been reported by photocatalysis, although this report was not with wastewater, it suggests that photocatalysis can also be a route to upgrade nitrogen-rich wastewater into ammonia [23].

Formic acid is also an important chemical recognized by being a hydrogen carrier or fuel. It is typically found in wastewater from chemical manufacturing, leather tanning and textile processing industry [24]. It is also a byproduct in wastewater treatment by advanced oxidation processes. This opens the window to look for the most selective AOP instead of only searching for the most efficient in terms of “burning” all organic matter. Formic acid was successfully synthesized from wastewater by a combined electrochemical-photochemical process [25]. Thus, this demands not only new experimental studies, but insightful and comprehensive revision of literature dedicated to wastewater treatment, with special emphasis in those processes where carboxylic acids were identified at some point in the process. Formic acid has also been synthesized by photocatalyzed reduction of CO_2 in stirred tank reactors and short reaction time reactors [26]. This type of reactor is promising to conduct selective oxidation and reduction reactions because of its excellent intrinsic mass transfer characteristics and short reaction times.

Thus, wastewater upgrading to value-added chemicals by any of the above-mentioned methods, other than biological and microbial fuel cells, is still in its infancy and its advance requires low cost, stable and selective electrodes, photoelectrodes and photocatalysts with narrow bandgaps and high light absorption, as well as an efficient products recovery process and optimized electrochemical cells and photoreactors. For the sake of water-energy-carbon nexus neutrality, natural sunlight-driven processes must be developed and optimized.

There is an important advance in hydrogen and methane production by algas, microbial, photocatalytic, electrochemical, photothermal, and photoelectrochemical processes. Leaving algas and microbial aside, the advances reached with the other methods have been mostly using synthetic water or CO_2 from a gas cylinder. To ease the extrapolation of these advances to the fields of wastewater recovery, first a complete characterization of the wastewater to be upgraded must be conducted. Its composition will determine not only the products to be upgraded but also the plausible technology to conduct it.

Data Availability Statement

Not applicable.

Funding

This work was supported by the SECIHTI Mexico under CVU 87755.

Conflicts of Interest

The author declares no conflicts of interest.

Ethical Approval and Consent to Participate

Not applicable.

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