



Are Wetlands as an Integrated Bioremediation System Applicable for the Treatment of Wastewater from Underground Coal Gasification Processes?

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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Abstract: Underground coal gasification (UCG) can be considered as one of the clean coal technologies. During the process, the gas of industrial value is produced, which can be used to produce heat and electricity, liquid fuels or can replace natural gas in chemistry. However, UCG does carry some environmental risks, mainly related to potential negative impacts on surface and groundwater. Wastewater and sludge from UCG contain significant amounts of aliphatic and aromatic hydrocarbons, phenols, ammonia, cyanides and hazardous metals such as arsenic. This complicated matrix containing high concentrations of hazardous pollutants is similar to wastewater from the coke industry and, similarly to them, requires complex mechanical, chemical and biological treatment. The focus of the review is to explain how the wetlands systems, described as one of bioremediation methods, work and whether these systems are suitable for removing organic and inorganic contaminants from heavily contaminated industrial wastewater, of which underground coal gasification wastewater is a particularly challenging example. Wetlands appear to be suitable systems for the treatment of UCG wastewater and can provide the benefits of nature-based solutions. This review explains the principles of constructed wetlands (CWs) and provides examples of industrial wastewater treated by various wetland systems along with their operating principles. In addition, the physicochemical characteristics of the wastewater from different coal gasifications under various conditions, obtained from UCG's own experiments, are presented.

Keywords: wetlands; bioremediation; bacteria; underground coal gasification (UCG); industrial wastewater

1. Introduction

Underground coal gasification (UCG) can be considered as one of the clean coal technologies that enables recovering valuable gas from coal in situ, even in the case of coal seams that are too deep, low grade or non-mineable with conventional methods. Produced gas can be applied in power generation, heat production or as a chemical feedstock. Years of development of UCG technology have shown that it is technically feasible and economically, socially and environmentally viable, i.e., due to its light surface footprint. The gasification process has come a long way from the first attempts in the former Soviet Union in the 1930s to complex plants with integrated combined cycle gasification or CO_2 capture and storage [1–5]. However, the UCG process in situ also carries some environmental risks, related mainly to the potential negative impact on surface and groundwater. Many heterogeneous and homogenous reactions that take place in the oxidation, reduction and pyrolysis zones developed during gasification affect the release of UCG-related contaminants from coal tars and ashes produced in the volatilisation process.

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Conducting the process below the hydrostatic pressure prevents gas migration to the surrounding rock strata and can effectively control contaminants migration because the groundwater influx is towards the cavity, however, the main environmental risk is related to the removal of process condensates and drainage of the operating site during unloading to the surface for cleaning. Numerous organic and inorganic contaminants are formed both during different process stages and after process termination. Then, the UCG wastewater and sludge contain a significant amount of aliphatic and aromatic hydrocarbons, including benzene, polyaromatic hydrocarbons and phenols, but also ammonia, cyanides and hazardous metals [6,7].

Among various physical, physico-chemical and biological processes, biological treatments have been identified as suitable and nature-based methods for the clean-up of wastewater contaminated with various pollutants. Searching for novel approaches for using biological systems based on natural biochemical processes has been the main topic of research. A holistic approach to bioremediation has been developed, involving the activity of microorganisms and terrestrial and aquatic plant species in the biodegradation of hazardous compounds. Natural wetlands (NWs) and constructed wetlands (CWs) are the examples of low-cost, sustainable, easy-to-operate and ecofriendly options for the effective removal of various organic and inorganic compounds. They are designed as natural biofilters that simulate natural processes dependent on the interaction between media (such as gravel, pumice rock, soil), plants and associated microorganisms. In particular, microbes are considered to be a key player in the removal of pollutants from contaminated wastewater [8]. Knowledge of the wetland microbial communities is useful in monitoring the restoration and wastewater treatment processes. Wetland ecosystems are habitats for all kinds of microbes, aerobic, anaerobic, anoxic and facultative. Their role is also crucial in the functioning and growth of plant communities, and for maintaining the sustainability of wetland ecosystems.

The aim of this paper is to review the use and development of wetland systems and to identify the challenges and propose alternative methods for the biological treatment of industrial wastewater, including wastewater from UCG processes.

2. Bibliometric Analysis

The output for the review was collected from the bibliometric database Scopus on 8 June 2022. The search included only English-language articles and reviews from all available years. Since a search for the phrases "constructed wetlands" and "underground coal gasification" returned no results, it was conducted for scientific publications containing the phrases "constructed wetlands" and "industrial wastewater" in the title, abstract or keywords. The total number of publications found in the study area was 321. Detailed searching criteria are presented in Table 1.

Table 1. Criteria and results of filtering scientific publications in the Scopus database.

Criterium	Phrase	Number of Publications Identified
1 Title, abstract, keywords	"Constructed wetlands" and "Industrial wastewater"	603
2 Document type and language	Article, Review, English	462
3 Subject area	Environmental Science, Agriculture and Biological Sciences, Chemistry, Engineering, Chemical Engineering, Multidisciplinary, Immunology and Microbiology	321

The first publication on the subject, entitled "Constructed wetlands for wastewater treatment" by S.C. Reed [9] in Scopus dates from 1991. Over the three consecutive decades, there has been a marked increase in interest in the topic of wastewater treatment using wetlands (Figure 1). By mid-2022, 13 articles had already been published, confirming the relevance of the topic of this review [10].



Figure 1. Number of publications in the study area between 1991 and 2022 (based on Scopus, 8 June 2022).

3. Underground Coal Gasification

3.1. Recent Worldwide Experiments

Underground coal gasification technology has been the subject of numerous pilotscale studies and experiments in mining basins around the world for years. Large-scale UCG research programmes were initiated in the 1950s in the Soviet Union, in the 1970s and 1980s in the United States and in the 1980s, intensive research was launched in the People's Republic of China [4]. Currently, the development of UCG technology to enable its widespread and commercial application is underway almost all over the world (including China, India, UK, Slovakia, Poland) and some countries such as Australia, the UK, Canada, New Zealand and the USA are already introducing licensing rules in this area [11]. The summary of significant UCG in situ trials operated after the year 2000 is presented in Table 2.

Table 2. Worldwide significant UCG experiments in situ after year 2000 (from [4] with some modifications).

Country	UCG Site	Startup Year	Coal Type/Seam Depth and Average Thickness [m]	Gasifying Agent
	Chinchilla G1	2000	subbituminous/132/10	air
	Chinchilla G3	2007	subbituminous/132/10	air
A (1)	Chinchilla G4	2009	subbituminous/132/10	air
Australia	Chinchilla G5	2011	subbituminous/132/5.5	air, oxygen/steam
	Bloodwood Creek P1	2009	subbituminous/200/9	air, oxygen/steam
	Bloodwood Creek P2	2011	subbituminous/200/9	air
Canada	Swan Hills	2009-2011	high volatile bituminous/1400/4.5	oxygen/steam
	Xinwen	2000	high volatile coal/100/1.8	air/steam
China	Feichang	2001	bituminous/90/1.5	air
	Xiyang	2001	anthracite/190/6	air/steam

3.2. Experiments from Central Mining Institute (GIG)

In recent years, intensive experimental studies in the field of UCG using large-scale experimental simulations on artificial coal seams were conducted by the UCG research group from the Central Mining Institute (GIG) in Poland. The first trials of surface UCG experiments at GIG were designed and developed within the RFCS-funded project HUGE 1 (2007–2010). Simultaneously with the UCG trials in Poland, the EU-based Research Fund

for Coal and Steel (RFCS) programme provided funding to support further UCG research in Europe. The most important projects in the UCG area include, apart from the above-mentioned pioneering project HUGE 1 and HUGE 2 (2011–2014), the following projects: TOPS (2013–2016), COGAR (2013–2016), Coal2Gas (2015–2017), UCG&CO₂Storage (2009–2010), MEGAPlus (2018–2021) and the ongoing UCGWATERPlus.

After 2010, more advanced ex situ experimental units for UCG simulation were designed and built at GIG's Clean Coal Technology Centre, and the GIG team conducted more than a dozen underground gasifications of coals of different rank, from semi-anthracite to ortho-lignite, using various gasification agents and under different conditions (Table 3).

Table 3. General characteristics of the main UCG in situ and ex situ experiments conducted by the Central Mining Institute in years 2007–2021.

Origin of Coal	Type of Coal	Type of Experiment/ Installation Pressure *	Gasifying Agent	Experiment Duration [h]	Coal Gasified [kg]	Wastewater Produced [kg]	Wastewater Outflow [kg/kg Gasified Coal]	References
Experimental mine "Barbara" (I) (Poland)	subbituminous	in situ	oxygen/air	355	21,980	14,810	0.67	[12]
Experimental mine "Barbara" (II) (Poland)	subbituminous	in situ	oxygen/steam	142	5364	2960	0.55	[13]
Hard coal mine "Wieczorek" (Poland)	subbituminous	in situ	air/oxygen/CO ₂	60 days	230,500	d.n.a.	d.n.a.	[14]
Hard coal mine "Bobrek" (Poland)	bituminous	ex situ	oxygen	48	176.9	46.0	0.26	[15]
Hard coal mine "Ziemowit" (Poland)	subbituminous	ex situ	oxygen	48	164.2	130	0.79	[15]
Brown coal mine "Bełchatów" (Poland)	lignite	ex situ	oxygen	50	970.0	480	0.49	[16]
Hard coal mine "Bielszowice" (Poland)	bituminous	ex situ	oxygen/air/steam	73	145.0	79.6	0.55	[16]
Premogovnik Velenje (Slovenia)	meta-lignite	ex situ	oxygen	120	730.0	d.n.a.	d.n.a.	[17]
Premogovnik Velenje (Slovenia)	meta-lignite	ex situ/3.5 MPa	oxygen	72	591.0	d.n.a.	d.n.a.	[18,19]
Coal mine Oltenia (Romania)	ortho-lignite	ex situ	oxygen/steam	96	790.0	d.n.a.	d.n.a.	[17]
Coal mine Oltenia (Romania)	ortho-lignite	ex situ/1 MPa	oxygen	72	585.0	d.n.a.	d.n.a.	[18,19]
Hard coal mine "Piast" (Poland)	subbituminous	ex situ	oxygen	72	2300	521	0.23	[20]
Coal mine "Six Feet" (UK)	semi- anthracite	ex situ/2 MPa	oxygen/steam	96	436.1	46.5	0.11	[21]
Coal mine "Six Feet" (UK)	semi- anthracite	ex situ/4 MPa	oxygen/steam	96	455.5	38.6	0.08	[21]
Hard coal mine "Wesoła" (Poland)	bituminous	ex situ/2 MPa	oxygen/steam	96	504	67.3	0.13	[21]
Hard coal mine "Wesoła" (Poland)	bituminous	ex situ/4 MPa	oxygen/steam	96	530.2	55.2	0.10	[21]

* no information means pressure close to atmospheric; d.n.a.-data not available.

3.3. Characterisation of Wastewater from Underground Coal Gasification Process: Experience from GIG

Wastewater from UCG is an example of industrial wastewater with an extremely complex matrix, containing large quantities of organic and inorganic pollutants, and therefore, the possibility of applying CWs to neutralise such wastewater appears to be important for investigating the possibility of biological treatment. Four main water sources in the process can be indicated: a. static water resources from the coal seam, b. dynamic resources (groundwater infiltrating from surface into the reactor environment), c. processing water—chemically bonded water present in minerals in the coal seam, d. water steam used as an addition to the gasifier and contain in the supply air of the UCG reactor [6]. The risk of groundwater contamination can be reduced by selecting an appropriate location and operation method of the pilot UCG plant. Wastewater generated during UCG has a similar composition to coking wastewaters [22], and requires complex mechanical, chemical and biological treatment [23] due to the high content of organic and inorganic impurities. According to Smoliński et al. [15], a higher total amount of wastewater (as well as higher coal consumption rate) is generated in the underground gasification of lignite than in the case of hard coal. This is most likely due to the high moisture content in the lignites. UCG wastewater is produced mainly during the evaporation of coal, coal-pyrolysis, gas cooling and gas purification processes. The wet UCG gas consists of water vapour, originating principally from the evaporation of coal moisture, pyrogenic water and from unintended hydrogen combustion. The amount of wastewater arising from UCG ought to be more minor than from traditional mining, nonetheless, the water flux from a gas scrubber includes a high concentration of hazardous compounds, e.g., phenols, PAHs, monoaromatic compounds (BTEX) and heavy metals, for which the concentration should be continuously monitored to avoid any leakage into the environment [24].

Total amount of wastewater produced during UCG experiments conducted in GIG is presented in Table 3. It should be noted that the total volume of UCG wastewater produced during in situ trials was difficult to evaluate, e.g., in the experimental mine "Barbara", approximately 500 kg of post-processing water per day was collected [25]. During ex situ experimental trials, the volume of wastewater obtained varied from 38.6 kg (gasification of semi-anthracite) to 521 kg (subbituminous coal "Piast") and 480 kg (lignite "Bełchatów"). If bed water is most responsible for the amount of wastewater, then gasification of lignite should produce more waste than, e.g., anthracite, which has the lowest moisture content among all coal types. It should be emphasised that the quality and quantity of the UCG wastewater obtained differs depending on many complex aspects of operating conditions, such as UCG experimental installation and process parameters. The volume and composition of UCG wastewater solidly depends on the type and rank of gasified coal and its properties, the parameters of the process (temperature and pressure), the type of gasifying agent and applied gasification technology. The amount of physicochemical compounds depends mainly on the thermodynamic conditions (temperature) and mutual proportions of pyrolysis and oxidation zones [18]. Moreover, the concentration of pollutants in UCG wastewater vary significantly with the progress of the UCG process. The average amount of wastewater produced was about 0.30 kg per 1 kg of the gasified coal. The relatively high wastewater production during gasification of bituminous coal "Bielszowice" in reference to the coal consumption (0.55 kg wastewater per 1 kg of coal) can result from the unreacted steam, the evaporation of water or hydrogen combustion [16].

The average values of the parameters determined in the UCG wastewater originating from in situ and ex situ experiments are presented in Table 4. The analyses were performed in the laboratories of the Central Mining Institute in Katowice; some of the results have been published [12,14,16–19,21] and some are included in unpublished reports. As it can be concluded from Table 4, the values of measured parameters significantly differ for each UCG experiment. The physicochemical parameters as pH range from 4.9 to 7.8, COD_{Cr} from 48 to 5060 mg O_2/L and BOD₅ from 300 to 4373 mg O_2/L . The ammonia nitrogen concentration varies from 11 to 7800 mg N/L, which might be determined by the pH values, and the sulphates level is between 33 and 3220 mg/L. In all wastewaters, low concentration levels of cyanides from 0.5 to 5.7 mg/L were observed. Additionally, the concentrations of metals were relatively low for all wastewaters, except iron which differs from 0.02 to 650 mg/L. Pankiewicz-Sperka et al. [21] validated that the type of gasified coal has a significant influence on the concentration levels of organic parameters. Moreover, metal concentrations occurring in raw coals have no direct impact on the composition of UCG wastewater, but it is rather driven by organic contaminants derived from the tars produced during the UCG process [21].

		Coal Type and Origin (Installation Pressure)											
Parameter/ Compound *	Unit	SB Barbara I (atm) [12]	SB Wieczorek (atm) [14]	B Bielszowice (atm) [16]	L Bełchatów (atm) [16]	SA Six Feet (2 MPa) [21]	SA Six Feet (4 MPa) [21]	B Wesoła (2 MPa) [21]	B Wesoła (4 MPa) [21]	ML Velenje (atm) [17]	ML Velenje (3.5 MPa) [18,19]	OL Oltenia (atm) [17]	OL Oltenia (1 MPa) [18,19]
pН	-	6.3	7.3	7.8	5.4	6.4	5.2	5.3	4.9	7.3	6.0	7.7	5.1
Conductivity	µS/cm	14,425	57,400	19,200	8638.0	1228.4	253.38	942.00	1006.7	2478.0	1770.0	3155.0	5253.0
COD _{Cr}	mg/LO_2	4308	5330	n.d.	n.d.	151.6	48.63	322.7	185.9	5060	691.0	2010	4177
BOD ₅	mg/LO_2	2228	2840	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	4373	300.0	1048	2105
Ammonia	mg/L N	1950	7800	3300	1225	160.1	11.68	96.41	95.74	280.0	189.0	463.0	778.0
Chlorides	mg/L	1660	18,000	n d	n d	11 15	11.68	29.18	45 94	n d	n d	n d	n d
Cvanides	mg/L	1 26	3 90	5 69	<0.5	1 11	1 43	1 70	0.87	1.31	0.70	1 01	3.00
Sulphates	mg/L	3220	980.0	651.0	1014	33.51	47.66	42.86	52 97	45 70	105.0	44.30	204.0
Mn	mg/L	4 91	n d	n d	nd	0.017	0.021	0.018	0.012	0.010	0.13	0.050	0.34
Fe	mg/L	650	n d	10.6	325	0.823	0.284	0 131	0.245	0.050	2 49	0.020	21.98
Sh	mg/L	<0.05	n d	n d	n d	0.036	0.12	0.064	0.013	0.030	0.030	0.030	0.070
As	mg/L	2 93	n d	n d	n d	0.036	<0.02	<0.01	<0.01	0.040	<0.005	0.21	0.16
B	$m\sigma/L$	6.5	nd	31	0.18	0.072	0.056	0.13	0.25	0.21	0.58	0.040	0.48
Cr	mg/L	0.51	7.3	0.022	<0.005	0.013	0.012	0.010	0.006	<0.005	0.17	<0.005	18
Zn	mg/L	3.53	0.570	1.15	10.8	0.021	0.499	0.320	0.200	0.060	0.180	0.080	0.300
Al	$m\sigma/L$	177	nd	nd	nd	0.031	0.046	0.029	0.023	0.060	0.220	<0.01	1 41
Cd	mg/L	<0.02	n d	n d	<0.002	<0.001	0.001	<0.0005	<0.0005	<0.000	0.001	<0.01	0.002
Co	mg/L	0.031	n d	n d	n d	0.004	0.003	<0.003	<0.003	<0.005	0.010	<0.005	0.043
Cu	mg/L	<0.01	0.062	0.065	<0.01	0.005	0.010	0.009	0.002	0.010	<0.005	0.010	<0.005
Mo	mg/L	0 133	n d	0 140	<0.01	0.005	<0.005	0.026	<0.005	<0.01	0.020	<0.01	0.011
Ni	mg/L	0.243	1 16	0.029	<0.01	0.098	0.312	0.051	0.027	0.050	1 16	0.010	2.83
Ph	mg/L	0.044	0.035	<0.02	<0.005	<0.005	0.064	0.046	0.060	0.010	0.040	<0.01	0.28
Hø	mg/L	< 0.005	n.d.	n.d.	n.d.	< 0.0005	< 0.0005	< 0.0005	< 0.0005	< 0.0005	< 0.0005	< 0.0005	0.003
Se	$m\sigma/L$	0.14	nd	nd	nd	0.016	0.017	0.036	0.027	0.040	<0.01	0.030	0.066
Ti	mg/L	0.52	n d	0.050	<0.005	<0.0005	0.001	0.001	<0.0005	<0.003	0.004	<0.003	0.055
Total	mg/L	484	820 **	3090	247	29.7	2.14	49.5	29.2	733	17.0	246	201
TOC	mg/L	616.0	1500	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2400	167.0	882.5	1250
Total BTEX incl.:	µg/L	55.80	414.0	790.0	106.0	5483	1497	2514	1354	1994	804.0	1784	1562
Benzene	ug/L	51.10	n.d.	504.0	96.00	4156	1341	2197	1059	1189	512.3	1190	1072
Toluene	$\mu g/L$	3.730	n.d.	140.0	7.000	n.d.	n.d.	n.d.	n.d.	356.3	175.2	277.0	236.3
Ethylbenzene	$\frac{1}{10}$	0.700	n.d.	22.00	0.500	n.d.	n.d.	n.d.	n.d.	238.7	24.40	263.5	209.8
Xvlene	це/L	1.820	n.d.	124.0	2.500	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Total PAH	μg/L	1912	399.0	1887	1066	1658	362.0	1090	407.2	n.d.	n.d.	n.d.	n.d.

Table 4. Composition of the UCG post-processing wastewater from selected experiments conducted in the Central Mining Institute.

n.d.—not determined, SB—subbituminous, B—bituminous, L—lignite, ML—meta-lignite, OL—ortho-lignite, SA—semi-anthracite. * In case of some parameters (e.g., metals), the limit of quantification of the method may be modified according to the presence of interfering substances. ** Total phenols volatile.

4. Bioremediation Process: Current Challenges and Trends

Bioremediation is a biological process using metabolic activities of living organisms, e.g., bacteria, yeast, fungi and plants, to destroy or neutralise to less hazardous organic and inorganic contaminants via complete mineralisation or co-metabolism. Compared to physico-chemical methods, generally, bioremediation technology is considered as less expensive, environmental friendly and more socially acceptable [26,27]. Most bioremediation systems are performed under aerobic conditions, but the systems under anaerobic conditions or aerobic–anaerobic conditions are also developed. In hybrid anaerobic–aerobic systems, recalcitrant pollutants are degraded by microbes in an oxygen deficit [28,29].

Two types of bioremediation methods are widely described in the literature, e.g., in situ bioremediation (bioventing, biosparging) and ex situ bioremediation (landfarming and composting, bioreactors, biopiles) [30].

The microorganisms involved in the bioremediation processes may come from a contaminated area (indigenous, autochthonus microflora) or they may be isolated from different places and inoculated to the contaminated site (allochtonus microflora). The supplementation of the microbial population is very effective where the native soil microorganisms are unable to degrade the pollutants. Introducing microorganisms to a contaminated site to enhance the bioremediation process is known as bioaugmentation [31,32]. The application of natural microbial consortia as inocula is a promising bioremediation method to remove anthropogenic compounds from polluted environments.

An option for bioaugmentation is biostimulation. The indigenous microorganisms are stimulated to grow and become active by changing environmental parameters such as temperature, pH or by adding nutrients, oxygen or surfactants [31,32].

Most of the organic hazardous compounds can be destroyed by the biodegradation process which is often a result of the actions of microbial consortium [33]. Microbes forming a consortium can perform multiple functions in bioremediation processes compared to a single microbe. Microbial consortia have great biotechnological potential for applications in long-term biodegradation. The application of microbial consortia is beneficial because of the synergistic interactions between different microbes that contribute to the overall contaminants degradation [34]. Microorganisms have the ability to degrade the contaminants from the enzymatic system into by-products that can be taken up by other microorganisms, which break them down into less toxic forms and harmless compounds that can be introduced into the biogeochemical cycles. In addition, different processes mediated by microbes, i.e., biomineralisation, biosorption and biodegradation, are part of the bioremediation process. Recently, research has focused on the design of microbial consortia, natural consortia or artificial co-culture systems as new emerging tools for bioremediation technologies. Dvorak et al. [35] presented the development of bioremediation technologies from the use of naturally occurring microorganisms called the era of Bioreme*diation 1.0* through the application of recombinant DNA technology that provides changes in the metabolic pathways of selected microbial hosts and the development of a patchwork strategy called *Bioremediation 2.0* to the era of new generations of bioengineering called Bioremediation 3.0. These novel engineering strategies combine high throughput technologies, computational methods and "omics" techniques to design and develop microbial biodegradation pathways to plan and execute an effective bioremediation process.

Various types of bioremediation technologies have been described in the literature to minimise the environmental pollution caused by xenobiotics of an anthropogenic origin, such as petroleum hydrocarbons, solvents, pesticides, dyes, antibiotics, heavy metals, explosives such as 2,4,6-trinitrotoluene and other chemicals [36–48]. Most of these compounds belong to the emerging contaminants, i.e., substances that remain in the environment for a long time [8]. Most scientists and researchers note that no single bioremediation method can clean up a contaminated environment because most contaminants occur in the form of mixtures containing organic and inorganic pollutants. In such cases, an effective bioremediation strategy should include the implementation of two or more methods [48,49].

Nowadays, the integration of bioremediation approaches is very popular and researches pay attention to the development of hybrid (synergistic) processes. Wetlands are now considered as a holistic and integrated approach using natural physical, chemical and biological processes such as sorption, precipitation, transpiration, sedimentation, plant activities, such as phytostabilisation, phytoextraction, evaporation, and biodegradation activities performed by associated microbial consortia [50]. Wetland systems are naturebased solutions (also popularly known as green solutions) and are a relatively new area of research. As nature-based solutions, they can provide many different services with high social, economic and environmental values, such as recreation, CO₂ sequestration, coastal protection, groundwater and soil moisture regulation, human well-being, flood regulation, biodiversity support and other benefits, such as the creation of new jobs, etc.

Next to the wetlands, algae-based technology named phycoremediation also belongs to nature-based solutions of wastewater treatment. It has been developed as a technique using algae for treating chemically contaminated water [51,52]. Algae have an ability to assimilate various toxic pollutants such as aromatic hydrocarbons, heavy metals and organochlorine [53,54]. Algae from various non-pathogenic genus such as *Chlorella*, *Spirulina* and *Scenedesmus* have been applied in the phycoremediation of phenolic compounds [55].

Municipal wastewater and industrial wastewater from paper, textile, tannery industries, petroleum refineries, agriculture wastewater, mining wastewater, stormwater and landfill leachate are only a few examples of polluted wastewater that has been treated by different kind of wetlands [56–59]. The application of wetlands for the purification of contaminated wastewater is rapidly increasing in the industrial sector.

5. Wetlands as Natural and Engineering Systems to Clean up Industrial Wastewater

Constructed wetlands (CWs) are defined as engineered systems whose operating principle is mainly based on natural ecosystem processes related to the decomposition and circulation of matter. These facilities use heterotrophic microorganisms and aquatic and hydrophytic plants to treat wastewater. The plants grow on appropriately designed objects, such as ground-root filters or ponds [60,61], which are designed to intensify and direct water purification. An important feature of CWs is that they are a low-cost, non-invasive, multifunctional and, above all, environmentally friendly solution [62]. The multifunctionality of CWs stems from their "natural character", whereby they blend into the landscape and, similar to their natural counterparts, can serve as urban wildlife refuge, recreational facilities and landscape engineering facilities [63].

In CWs, by creating conditions that allow the growth of hydrophytes, it is possible to intensify the redox processes which, supported by other processes such as sorption, sedimentation and assimilation, allow the removal of significant amounts of pollutants from wastewater. Plants used in these systems such as *Phragmites australis* often form an extensive system of rhizomes and roots around which, due to internal gas transport pathways [64], local oxygen microspheres are formed, surrounded successively by hypoxic and further reductive microspheres [65]. The occurrence of aerobic and anaerobic zones around the roots stimulates the decomposition of organic matter and the nitrification process, occurring thanks to the metabolic activity of microorganisms-bacteria, algae, fungi and protozoa. Another desirable feature of plants used in CWs is the rapid growth of biomass, which, among other advantages, has a beneficial effect on the removal of nutrients from treated waters. To create CW systems, naturally occurring wetland plants are used, which can include: emergent plants (Phragmites australis, Typha latifolia, Typha angustifolia and Salix viminalis), submerged vegetation (Hydrilla verticillata, Ceratophyllum demersum, Vallisneria natans, Myriophyllum verticillatum and Potamogeton crispus), free-floating plants (Nymphaea tetragona, Nymphoides peltata, Trapa bispinosa and Marsilea quadrifolia) and floating leaves plants (Eichhornia crassipes, Salvinia natans, Hydrocharis dubia and Lemna spp.).

Constructed wetlands have become very popular in recent years as an alternative to high-tech wastewater treatment solutions [59,62,66]. These systems are suitable for treating wastewater from both point and area sources [67,68]. Due to their ability to assimilate

nutrients, CWs have great potential in treating municipal wastewater. However, it is increasingly observed that these solutions are used to treat industrial wastewater and leachate [62,69,70]. CWs treat a wide range of pollutants, such as heavy metals, pesticides, petroleum hydrocarbons, explosives, radionuclides and pollutants specific to effluents from a textile dye factory, etc. Table 5 lists experiments using CWs, including small-laboratory-scale systems that have been used to remove various compounds.

CWs can be divided into two types, those operating on a surface flow system (FWS, free water surface) and those operating on a subsurface flow system. FWS treatment plants function as ponds or canals that usually provide a serpentine water flow. In FWS treatment plants, the long retention time (usually between a few days and 2 weeks) and large surface area favour the removal of solids and organic matter [71]. Different types of vegetations are used in this type of treatment plant, but it is worth noting that in Europe, the most commonly used macrophyte is *Phragmites australis* [72,73].

Subsurface flow plants are constructed so the wastewater flows under a layer of material filling the bed. In subsurface flow CWs, the retention times are usually shorter (1–2 days) and active microorganisms are associated with plant root systems and the substrate surface [74]. There are two types of subsurface flow CWs: with horizontal flow (HSF, horizontal subsurface flow) and with vertical flow (VSF, vertical subsurface flow) [75]. It also happens that mixed systems, with both a vertical and horizontal wastewater flow, are used in a single treatment process. The division of constructed wetlands is presented in Figure 2.



Figure 2. Simplified classification of constructed wetlands (CWs) for wastewater treatment (from [68] with some modifications); * FWS CWs may contain different types of vegetation, such as emergent, submerged, free floating and floating-leaved plants; ** VSF CWs can be characterised by different wastewater flows: up, down and tidal.

In summary, CWs in various configurations can deal with a whole group of different types of pollutants found in industrial wastewater (Table 5). Considering that many of them, such as hydrocarbons, cyanides, phenols and some heavy metals, are present in UCG wastewater, it is reasonable to assume that a properly designed CW should be able to treat UCG wastewater. However, the literature suggests that in the case of high PAH levels, which often occur after the UCG process (Table 4), it would be advisable to aerate the system and extend the hydraulic retention time (HRT) accordingly.

Pollutants	Type of Wastewater	Type of Wetlands and the Plant Species Used	Removal Rate/ Comment	References
Heavy metals (Mn, Cd, Zn and Pb), arsenic (As)	Mining (the pilot-scale experiment)	The combination of adsorption (modified iron-ore drainage sludge) and HSF CWs with <i>Phragmites australis</i>	The average removals during four months of operation was as follows:Mn—96,9%, Cd—79,6%, Zn—52,9%, Pb—38,7% and As—96,9%	[76]
Heavy metals (Fe, Mn, Al, Co, Ni and Cr), sulphate (900–1500 mg L ⁻¹)	Synthetic acid mine drainage (laboratory-scale experiment)	HSF-CWs planted with <i>Typha latifolia</i> and organic-rich substrate (cow manure and bamboo chips) Natural wetland	After the 6-month metal removal efficiency: Cr—99.7%, Ni—97.8%, Co—93.7%, Fe—91.6% and Al—59.7%. Microbial sulphate reduction 44–75%.	[77]
Heavy metals (Mn, Cu, Co, Cr and Cd)	Coke plant effluents	Dominant emergent plants: Colocasia esculenta, Scirpus grossus and Typha latifolia	Natural wetlands seems to be efficient in removal of selected heavy metals from coke-oven effluent	[78]
Heavy metals, phenol	Post-methanated distillery effluent (PMDE)	FWS CWs in India planted with <i>Typha</i> angustata L.	Removal: Cd (34–62), Cr (36–58), Cu (33–54), Fe (33–52), Mn (36–83), Ni (36–59), Pb (33–60), Zn (32–54), phenol—93.75% after 7 days of free water surface flow treatment	[79]
Ammonium, iron and traces of organic compounds	Coke plant effluents(pilot-scale)	HSF-CWs with two-stage gravel bed planted with <i>Phragmites australis</i>	Nitrogen removal efficiency (54–94%). Removal of COD from 35 to 52% of inlet concentrations	[80]
Heavy metals (Cu, Ni, Pb and Zn), cyanides (CN ^{$-$}) and sulphates (SO ₄ ^{2–})	Synthetic electroplating wastewater (laboratory-scale experiment in columns)	Up-flow VSF-CWs (lactates as carbon source, peat or gravel as medium). Some columns planted with <i>Phragmites</i> <i>australis</i>	Maximum removal: Cu, Ni, Zn, $CN^- > 90\%$; Pb > 70% SO ₄ ²⁻ > 10%. Insignificant effect of vegetation	[81]
Phenol, m-cresol, methyl tertiary-butyl ether (MTBE), benzene	Contaminated groundwater (the pilot-scale experiment)	Three separate HSF CWs. Two of them with <i>Phragmites australis</i> and one without vegetation	Surface load removal rates (SRR; mg m ⁻² d ⁻¹) were as follows: MTBE—20, m-cresol—80, benzene—335, phenol—620. The presence of <i>Phragmites australis</i> significant improved the contaminant removal performance.	[82]
Phenol, ammonium andnitrogen	Domestic wastewater spiked with phenol (laboratory-scale experiment)	HSF CWs planted with <i>Typha latifolia</i> with different substrate	remove phenol completely ($C_0 = 500 \text{ mg}$ L^{-1}) after 36 days with rice husk in substrate and by 60% in gravel in substrate. Planted wetland units performed better than the unplanted ones.	[83]
Phenol, organics (COD), thiocyanate and ammonium nitrogen	Synthetic wastewater (laboratory-scale experiment)	HSF-CWs planted with Typha angustifolia	Efficiency removals (operation time period—158 days): Phenol- 99%, COD—93%, ammonia nitrogen—17–30%. Alkalinity improved thiocyanate removal to 91%.	[84]
BTEX	the former refinery site(pilot-scale system consisted of four subsurface flow treatment cells equipped with aeration).	HSF-CWs. Types of plants: Salix, Phragmites, Scirpus, Juncus and Cornus	Removal after one year operating: benzene—80%, total BTEX- 88%	[85]

Table 5. Examples of the experiments described in the literature using wetlands as a bioremediation approach.

Pollutants	Type of Wastewater	Type of Wetlands and the Plant Species Used	Removal Rate/ Comment	References
PAHs (naphthalene; mixture of phenanthrene and pyrene)	A laboratory-scale experiment investigating the effects of PAHs on plant growth and development.	Hydroponics/pot experiment.Species: Baumea juncea, Baumea articulata, Schoenoplectus Validus and Juncus subsecundus	The effect of PAHs on plant growth in CWs may be species-specific and can depend on the type of PAHs and the substrate	[86]
PAHs	A laboratory-scale experiment	VSF-CW planted with Iris pseudacorus	The reduction of phenanthrene using biochar-loading copper ions (Cu-BC) was about 94% (HRT lasted 3 days)	[87]
Hydrocarbons and cyanides		FWS-CWs. Shallow basins, initially planted with: <i>Typha latifolia</i> and <i>Schoenoplectus</i> <i>tabernaemontani,</i> subsequently converted to <i>Ceratophyllum</i> <i>demersum</i> and <i>Potamageton spp</i> .	Removal after 7 days detention: total cyanide—56%, free cyanide—88%, Gasoline, diesel—~67%	[88]

Table 5. Cont.

6. Role of Microorganisms in Wetlands

Wetland ecosystems are examples of reservoirs of microbial diversity, mainly bacteria, fungi and actinomycetes [89]. In wetland, most biogeochemical activities are carried out by microbes. In particular, bacteria are the most active in processes occurring in wetland nutrient cycling and biodegradation processes because of their specific properties such as small genome size, short replication time, rapid evolution, relative simplicity of structure, metabolic properties and adaptation to new and extreme environmental conditions. It is known that microbial communities occurring in wetlands intensively enhance various processes. Investigating the interactions of the microbial structure and functions with wetland plants is an important part of specific transformations, biodegradation, biogeochemical cycles (including nitrogen, carbon, phosphorus and sulphur cycles), wetland survival and restoration. Many processes of nitrification, denitrification, mineralisation, humification and absorption occur through physical, chemical and microbial activity. Processes mediated by microorganisms are of great importance for the bioremediation function of wetlands. In wetland, aerobic and anaerobic microbial activity and microbial communities are mainly established by plant roots. Literature reviews by Liu et al. [90] and Wang et al. [91] show that some parameters such as temperature, redox potential and dissolved oxygen (DO) are influential factors for pollutants' degradation in wetland systems. DO is a vital factor that could influence microbial activities and the efficiency of pollutants' removal. Special attention is given to the mechanism of radial oxygen loss (ROL), that is responsible for contaminant removal in wetlands and the activity of the root-associated microbiome (rhizobiome).

The first research papers characterising various types of wetlands (such as rice paddies, acidic wetlands—peatlands, freshwater wetlands, black mangroves, salt marshes, boreal wetlands) and their microbes were published in the 21st century [92–100]. These papers have mainly focused on the characterisation of a specific group of bacteria involved in the nutrient cycles. Current knowledge on the diversity and functioning of wetland microbial communities is insufficient and there are many gaps that need to be assessed in future wetland studies. The characterisation of wetland microbial communities is fundamental for understanding the working rules of such a complex system as a wetland ecosystem. Up to now, phenomena and principles of microbes' activities and plant–microbial interactions in wetlands are priorities in the development of bioremediation research strategies [101].

The synergistic effects of microorganisms and plants have been applied to develop bioremediation processes [102]. With respect to remediation purposes, root-associated

bacteria (rhizobacteria) are considered most relevant. They form the area surrounding plant roots, called the rhizosphere, that is influenced by the growth, respiration and secretions of the plant roots. Most of the bacteria adhere to the root surface and form the microbial biofilm which is named the rhizoplane. The root-associated microbial community, called the rhizomicrobiome, contains around 10¹¹ microbial cells per gram of root tissue and more than 30,000 prokaryotic species [103]. Most of the root-associated bacteria belong to plant growth-promoted bacteria (PGPB) which enhance plant growth by the secretion of specific phytohormones such as siderophores, aminocyclopropane-1-carboxylase (ACC) and indole acetic acid (IAA). The enzyme ACC deaminase is known as an ageing hormone and for the improvement of plant nutrient uptake through nitrogen fixation and phosphate solubilisation [104]. Recently, the beneficial role of rhizobacteria in the promotion of plant growth and pollutant removal has been reviewed in detail by Orozco-Mosqueda et al. [105]. Rhizobacteria also produce various metabolites such as antibiotics, biosurfactants, enzymes, cellulases and HCN. The role of bacterial biosurfactants in environmental bioremediation processes has gained importance due to their low toxicity, high biodegradability and ecological acceptability. They are excellent emulsifiers and foaming and dispersing agents due to their surface activity. Biosurfactants such as rhamnolipids, sophorolipids and surfactins can effectively solubilise, emulsify and mobilise both heavy metals and organic pollutants. Biosurfactants are also known as antimicrobial agents inhibiting the growth of bacterial and fungal pathogens. Nowadays, interest in the plant root microbiota, the relative metabolites, and its activity is rapidly growing. However, there are still a huge number of cultivable and uncultivable microbes whose ecology and biochemistry are unknown. Recently, the research on changes in the structure and function of microbial communities has been developed by the advancement in science and technology, especially in the field of molecular techniques [106–108]. There are two main types of methods used to assess the structural and functional diversity of wetland microbes, i.e., culture-dependent and cultureindependent methods. Culture-dependent methods provide the detection, enumeration and determination of physiological profiles of microbial species using traditional plate count methods, while culture-independent molecular approaches are used to characterise the unculturable fractions of wetland microorganisms [109]. Figure 3 shows a flowchart of the methods used to characterise the functional and structural diversity of wetland microbial species and communities.





Wetland communities are characterised by changes to the community composition (structure) and by assessing changes in community function. The 16S rDNA gene as a target is currently used for the taxonomic classification of microbes. In contrast, the community function can be assessed by microbial enzyme activity, the degradation of various substrates or evaluation of functional genes. The most popular method to evaluate the microbial or community function is community-level physiological profiling (CLPP), conducted using BiologTM EcoPlates. This method was used by Salomo et al. [110] to characterise the metabolic potential of a microbial community in a constructed wetland with a vertical flow. Jamwal and Shirin [111] conducted a laboratory-scale experiment to identify microflora using the traditional plate method in planted and unplanted constructed wetland systems. Authors focused on the characterisation of microbes from the rhizosphere region of Typha domingensis using traditional culture methods, e.g., serial dilution and plate methods. Bacterial strains involved in the nutrient cycles and playing a significant role in the degradation of organic compounds such as *Pseudomonas* spp., *Bacillus* spp., Staphyococcus spp., Corynobacterium spp., Streptococcus spp., Lactobacillus spp. and Proteus spp. were isolated and characterised. In contrast, Xu et al. [112] combined metagenomic analysis and enzyme activity measurements to evaluate the distribution of the bacterial community in a vertical flow constructed wetland (VFCW) system. Proteobacteria were the most abundant phyla, followed by Bacteroides, Firmicutes, Acidobacteria and Chloroflexi. However, about 10% of unknown bacteria were also detected. The results showed that there was no relationship between enzyme activity and microbial populations. Some relations were observed between the functional bacterial community and the activity of particular enzymes, e.g., between Nitrospira and urease. Dynamic changes in enzyme activity were observed in different layers of CWs. In the paper of Xiang et al. [113], a constructed wetland system with Acorus calamus was established to investigate the bioremediation process of petroleum-contaminated wastewater. In this study, a high-throughput sequencing system was used to characterise bacterial diversity and the community structure. The results showed that the A. calamus root system forms a rhizosphere effect, which provides good conditions for the growth of specific microbes in the wetland. The dominant bacteria in the constructed wetland were as follows: Acinetobacter, Pseudomonas, Rhizobium and Rhodobacter. Pearson correlation analysis revealed a strong correlation between the values of richness and diversity indices and the removal efficiency of petroleum pollutants. The identified bacteria are involved in organic matter decomposition, nitrogen removal and the decontamination of petroleum pollutants. They have specific organic compound degradation properties such as biosurfactant production.

The study by Ma et al. [114] used Illumina high-throughput sequencing to identify the profile of microbial communities in wetlands constructed at a pilot-scale during saline wastewater treatment. Variations in the diversity and composition of microbial communities between the roots of *Salicornia bigelovii* and wetland layers were evaluated. Among twelve phyla detected in all samples, eleven were bacterial phyla and only one represented the archaeal phylum—*Thaumarchaeota*. Microbiological differences were observed between the root zones and the wetland layers. *Proteobacteria, Firmicutes, Cyanobacteria* and *Bacteroidetes* were dominated in the root zones, while *Cyanobacteria, Proteobacteria, Firmicutes, Verrucomicrobia* and *Bacteroidetes* were widespread in the wetland layers. These differences were probably related to the oxygen concentration and root secretions. Furthermore, the analysis indicated that specific functional genera such as *Nitrosopumilus, Vibrio, Pseudoalteromonas, Nitrospina* and *Planctomyces* were present in the wetland system. These play a key role in the promotion of the growth of wetland plants and in the removal of nitrogen and phosphorus pollutants.

The review article of Lv et al. [115] summarises the results of a study related to the search for 16S rRNA gene sequences from wetland ecosystems deposited in two public databases: GenBank and RDP. This meta-analysis showed that a total of 12677 bacterial and 1747 archaeal sequences from various wetland ecosystems were collected in the GenBank database in 2012. All bacterial sequences were assigned to 6383 operational taxonomic units (OTUs), representing 31 known bacterial phyla. The predominant bacterial phyla were: *Proteobacteria, Bacteroidetes, Acidobacteria, Firmicutes* and *Actinobacteria*. The genus *Flavobacterium* (11.6% of bacterial sequences) was the dominant bacterium in the wetland, followed by *Nitrosospira* and *Nitrosomonas*. Archaeal sequences were mostly assigned to the

Euryarchaeota and *Crenarchaeota* phyla. The dominate archaeal genera were *Fervidicoccus* and *Methanosaeta*.

The review by Mellado and Vera [107] focuses on the description of microbes detected in both natural and constructed wetlands that belong to the Bacteria and Archaea domains. Microorganisms were identified and characterised using a high-throughput, culture-independent sequencing technique. The 16S rRNA gene was used as a target to classify Bacteria and Archaea in wetlands. Functional genes have also been used as genetic markers to identify the specific metabolic groups of microbes. Among the bacteria, *Proteobacteria* was the most abundant phylum, both in natural and constructed wetlands. *Proteobacteria* members participate in most metabolic processes, such as the degradation of organic matter, organic compounds, biochemical cycles such as the methane nitrogen cycle, the sulphur and phosphorus cycle and methanogenic oxidation. The specific group of microorganisms perform non-conventional pathways such as heterotrophic nitrification, anammox and autotrophic denitrification to remove organics and nutrients [116–118]. Based on some recent reviews, there is an increasing research interest in studying microbial communities to track the success of constructed saltmarsh and wastewater treatment processes [119,120].

7. Conclusions

Wetland systems are an example of eco-industry which focuses on environmentally friendly clean-up technologies using the natural processes of plant-associated microorganisms. The valorisation of contaminated industrial wastewater, also in the context of clean coal technologies to which the UGC process belongs, appears to be one of the pillars of a modern and responsible industry. Wetlands have recently received a lot of interest from researchers in the treatment and removal of anthropogenic pollutants from wastewater as they are considered as nature-based solutions whose application would have a positive impact on the environment.

Despite this, the most important information about the operation and processes of wetlands is still unknown. Knowledge about the type of microorganisms and their role in designing efficient wetland systems for bioremediation processes is still needed.

This review article systematises the latest knowledge and recent developments in the treatment of industrial wastewater in wetland ecosystems, particularly in terms of the removal of organic and inorganic pollutants present in difficult matrix wastewaters, such as those from UCG processes. The review suggests that constructed wetlands could be a multi-green remediation approach for treating industrial effluents, including wastewater from coal gasification processes. It can be expected that properly designed CWs can purify wastewater from underground coal gasification, however, it should be noted that high concentrations of certain pollutants that often accompany UCG, e.g., PAHs, may require the additional aeration of the wetland system and the hydraulic retention time extension.

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