

Vacuum-UV Radiation at 185 nm for Disinfecting Water

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Abstract: The vacuum-UV (VUV) radiation of water leads to the in situ formation of hydroxyl radicals. Low-pressure mercury vapor lamps which emit at 185 nm are potential sources of VUV radiation. Zoschke et al.^[1] presented an excellent discussion of the utilization of VUV radiation at 185 nm for treating water comprising the conversion of inorganic and organic water constituents, and the disinfection performance. One more focal point stays on the production of ozone via VUV radiation from oxygen or air and the usage of the formed ozone in integration with VUV irradiation of water in the VUV/O₃ method. This work focuses on the merits and restriction of the VUV technology at 185 nm as well as likely usages in disinfecting water is outlined. Practically, VUV irradiation stays not a true stand by to traditional methods in water treatment, like adsorption on activated carbon, or to diverse advanced oxidation processes (AOPs) because of the inherent restrictions of the VUV technology. Nevertheless, VUV irradiation bids fresh potentials for particular utilization such as the provision of ultrapure water or as a principal treatment method for decentralized setups. Like for many AOPs, the most suitable use in (large-scale) water treatment remains the implementation as a pre-treatment to improve bio-decomposition.^[1]

Keywords: Vacuum-UV (VUV) irradiation; 185 nm; Low-pressure mercury vapor lamps (LPMVLs); Advanced oxidation processes (AOPs); Disinfection; Ozone generation

1. Introduction

Ultraviolet (UV) radiation includes the wavelength span from 1 to 380 nm. Inside such scope, it may be differentiated among UV-A (380-315 nm), UV-B (315-280 nm), UV-C (280-200 nm), vacuum-UV (VUV) (200-100 nm), and extreme UV (100-1 nm).^[1-2] For UV radiation, the most frequent sources remain low-pressure mercury vapor lamps (LPMVLs) with a partial pressure of mercury of around 1 Pa. Such pressure correlates with the vapor pressure of liquid mercury at 40°C at the lamp wall. LPMVLs show an efficiency of 25-45% in the emitted wavelength span. The emission spectrum of LPMVLs is concentrated at a restricted number of well-defined lines. Thus, the sources are named monochromatic. The large resonance lines are at 253.7 and 184.9 nm. The relative intensity of the emission for classical LPMVLs related to the most significant line at 254 nm (quoted as 100%) is illustrated in Table 1.

The 254 nm emission line is straight beneficial for killing pathogens. Thus, in the so-named germicidal lamps, the 185 nm line is removed via employing suitable glass materials like optical glass or quartz doped with titanium dioxide.^[3] The vacuum-UV emission at 185 nm may only be employed for irradiation when a synthetic quartz glass of high purity (Suprasil quartz) is utilized to cover the lamp.^[2-3] The tests and methods that employ such a lamp emitting at

254 and 185 nm are in this work alluded to as 'VUV' to distinguish them from the UV radiation at 254 nm.

A different lamp technique for the emission in the VUV span is Xe-excimer radiators which emit at 172 nm and are mostly used for advanced oxidation technologies.^[2] Additional data and instances for implementing Xe-excimer lamps may be read in many publications.^[2-7] Zoschke et al.^[1] concentrated on VUV irradiation at 185 nm emitted by LPMVLs.

Water highly absorbs light in the VUV domain.^[1] The absorption coefficient at 185 nm is $\alpha = 1.8 \text{ cm}^{-1}$.^[8] This agrees with a molar absorption coefficient of $\epsilon = 0.032 \text{ L/(mol cm)}$ (computed with a concentration of water of $c(\text{H}_2\text{O}) = 55.49 \text{ mol/L}$ at a density of 1 kg/L). The film in which 90% of the initial irradiation is absorbed is very thin and has a thickness of $l = 5.5 \text{ mm}$.^[9] Fig. 1 displays a comparative depiction of the absorption of UV and VUV irradiations.

2. Generation of hydroxyl radicals

As a result of the absorption of the VUV radiation, the homolysis (Eq. (1)) and photochemical ionization of water happen (Eq. (2)).^[1] The quantum yields, V , for these reactions are 0.33 and 0.045, respectively.^[6] The produced products are hydroxyl radicals, hydrogen atoms, and solvated electrons (Eqs. (1) and (2)) which

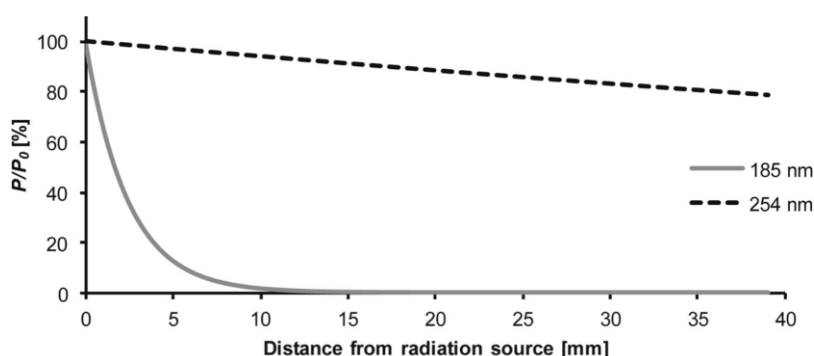
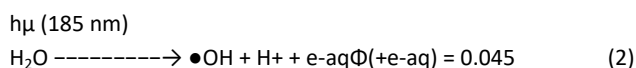
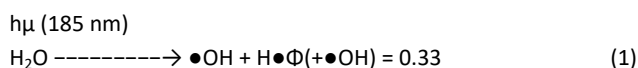


Fig. 1. Radiant power P at diverse distances from the irradiation source referred to the initial radiant power P_0 for water with a transmittance of 94% (data computed employing Lambert-Beer's Law).^[1]

Table 1. Emission intensity of LPMVLs relative to the emission at 254 nm.^[3]

| λ (nm) | Emitted intensity (I_0 , rel) |
|----------------|----------------------------------|
| 184.9 | 8 |
| 296.7 | 0.2 |
| 248.2 | 0.01 |
| 253.7 | 100 |
| 265.2-265.5 | 0.05 |
| 275.3 | 0.03 |
| 280.4 | 0.02 |
| 289.4 | 0.04 |
| 405.5-407.8 | 0.39 |
| 302.2-302.8 | 0.06 |
| 312.6-313.2 | 0.6 |
| 334.1 | 0.03 |
| 365.0-366.3 | 0.54 |

begin manifolds of reduction and oxidation reactions involving molecular oxygen and oxygenated species.^[2,6,10]



Because of the direct generation of hydroxyl radicals, VUV irradiation is considered between the advanced oxidation processes (AOPs).^[1,11-14] VUV photolysis of water is a greatly efficacious technique for forming advanced oxidation circumstances and the production rate of hydroxyl radicals is similar to remaining AOPs.^[15,16]

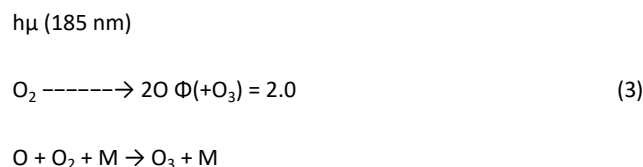
In 1996, researchers^[17] proved that the hydroxyl radicals were produced under VUV irradiation. During such a process, the hydroxyl radicals are converted into more stable radicals utilizing a spin-trapping agent. Because the hydroxyl radical concentration could not be measured, scientists^[17] contrasted the studied techniques via determining the height of the peaks. The peak height for the spin-trapping agent augmented in the order $\text{O}_3/\text{UV} < \text{O}_3/\text{VUV} \approx \text{VUV}$.^[1]

In 2009, the generation of hydroxyl radicals throughout VUV irradiation has as well been characterized by Kutschera et al.^[18] They controlled the decomposition of the probe compound par-chlorobenzoic acid (p-CBA) to measure the hydroxyl radical concentration. Such a procedure, which has previously been utilized for the AOPs UV/H₂O₂ and ozonation, is as well suitable for the irradiation at 185 nm. Nevertheless, awareness should be accorded

to the photolysis of p-CBA below UV and VUV light which was as well investigated by other researchers.^[9] Via VUV irradiation, Kutschera et al.^[18] regulated a hydroxyl radical production rate of 1.4×10^{-9} mol/L per 1 J/m² (fluence at 254 nm). The production of hydroxyl radicals is a function of the emission power of the lamp, the reactor design, and the optical characteristics of the irradiated medium.^[19] The measured $\bullet\text{OH}$ generation rate may be utilized to compute the steady-state hydroxyl radical level $[\bullet\text{OH}]_{ss}$, in a particular water matrix. The hydroxyl radical level in the setup, which impacts the decomposition of organic material, is influenced via the irradiated medium, mostly the sum of radical scavengers.^[1] With the $[\bullet\text{OH}]_{ss}$, the decomposition rate constant for microcontaminants which are mostly degraded via the indirect reaction with hydroxyl radicals may be divined, as illustrated by Kutschera et al.^[18] The indirect hydroxyl radical evaluation with p-CBA was also implemented by other researchers.^[20] They estimated time-based $\bullet\text{OH}$ exposure rates for the techniques VUV/O₃ and UV/O₃ (at 4 mg O₃ per mg C) of 1.2×10^{-11} and 4.0×10^{-12} mol \times min/L, respectively. Such amounts are matrix-specific and cannot be juxtaposed with the hydroxyl radical production rate suggested by Kutschera et al.^[18]

3. Production of ozone

In the gas phase, the VUV emission at 185 nm may be employed for the photochemical production of ozone from oxygen that can be used as an oxidizing agent in water treatment.^[2-21] The absorption of the VUV radiation via oxygen provokes the photolysis of the oxygen molecule. In the reaction of the oxygen atom with another oxygen molecule, ozone is produced (Eq. (3)). Such a reaction occurs in the existence of an extra molecule M which absorbs the excessive kinetic energy.^[1,2,22]



Because of the concomitant emission at 254 nm by the LPMVL, the photolysis of ozone at 254 nm happens beside the photochemical ozone formation (Eq. (4)).^[1,2,22]

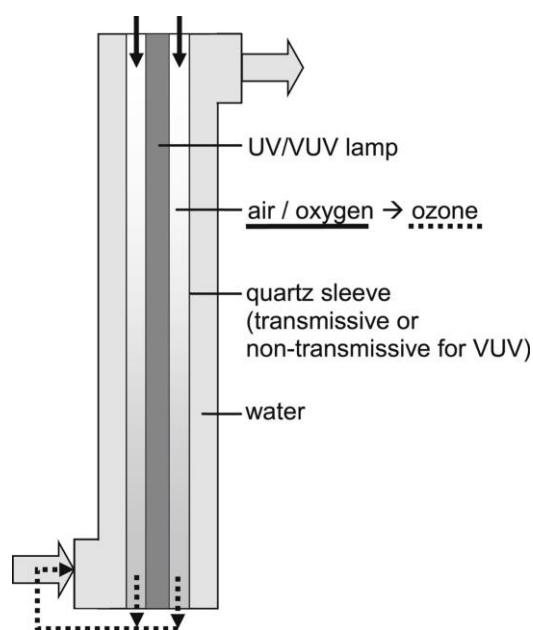
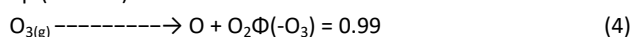


Fig. 2. Schematic cross-section of a device with internal ozone production.^[1]

$h\nu$ (254 nm)



The gas-phase absorbance coefficient (on a log scale) of oxygen at 185 nm is around $0.1 \text{ cm}^{-1} \text{ atm}^{-1}$ and much lower than the absorbance of ozone at 254 nm ($135 \text{ cm}^{-1} \text{ atm}^{-1}$).^[2-21] Besides, the relative emission intensity at 185 nm is lower than at 254 nm (see Table 1). Consequently, the photo stationary equilibrium of ozone generation vs. ozone degradation with traditional mercury vapor lamps is disadvantageous. Apart from the 254 nm UV light, the heat of the lamp and the existence of water vapor ameliorate the degradation of ozone.^[21] Nevertheless, employing ambient air for ozone formation provides the merit that no additional feed gas or equipment for the drying of air is required. A drawback of the ozone production via VUV irradiation is a fast aging of the quartz of the lamp provoking a loss of transmission at 185 nm wavelengths by at least 50% after 500-700 h operation time.^[1-3]

In 1959, Briner^[23] investigated the photochemical formation of ozone with mercury lamps and attained a maximum ozone yield of 1.8 g/kWh employing liquid oxygen and a 450 W lamp. A lower lamp power of 7W and oxygen gas yielded 0.3-0.4 g/kWh ozone and the ozone yield augmented with higher flow rates.^[23] In an investigation with LPMVLs, Dohan and Masschelein^[21] obtained ozone yields of 16-27 g/kWh and 7 g/kWh employing oxygen and dried air respectively. The ozone production at 172 nm with Xe-excimer sources was explored by Hashem et al.^[4] The attained ozone levels in the gas phase were 7.5 mg/L formed from oxygen and 6 mg/L from the air at a flow rate of 2 mL/min. This correlates with a yield of 9 and 7.2 g/kWh, respectively.^[1]

Bolton and Denkwicz^[22] presented a model to compute the ozone level formed by 185 nm VUV radiation. Such a model is founded on the absorption features, partial pressure, and quantum yields for ozone generation.^[1] The ozone levels divined by such a

model were bigger than the measured ozone level because of the thermal degradation of ozone near the lamp surface which was not involved in the model.^[22]

Regardless of the small ozone yields of LPMVLs, such radiation sources sustain the merit that the photochemical formation of ozone and the irradiation of water may be integrated into a reactor setup employing only one lamp for both techniques. A schematic cross-section of an annular irradiation reactor with internal ozone production is illustrated in Fig. 2. An additional illustration of an ozone-producing UV lamp is provided elsewhere.^[22] The VUV radiation at 185 nm is absorbed by oxygen in the air to form ozone. This reaction occurs in the annular space among the UV lamp and the quartz sleeve. The produced ozone enriched gas is injected into the water passing through the UV device on the other side of the quartz sleeve.^[22] Moreover, the 254 nm UV light or the 254 and 185 nm emission comes across straight via the quartz sleeve for the irradiation of the aqueous phase. The AOPs that may be employed with such technique are UV/O₃ or UV/VUV/O₃. Applying the two responses, ozone production in the gas phase and photolysis of an aqueous phase, through only one radiation source ameliorates the decomposition performance of organic contaminants.^[1-4]

UV setups with internal ozone production have been illustrated by Bolton and Denkwicz^[22] for killing pathogens targets, by Hashem et al.^[4] with a Xe-excimer source, and by the European Patent EP1394118^[24] using a non-electrode irradiation source emitting at 185 and 254 nm. Zoschke et al.^[25] employed a device with internal ozone production with LPMVL as shown in Fig. 2 for the decomposition of micro-contaminants.

4. Reactions of water ingredients

Zoschke et al.^[1] deeply discussed the transformation of inorganic water constituents, elimination of organic matter in potable and wastewater treatment, and decomposition of organic trace compounds. Therefore, more details about vacuum-UV Radiation at 185 nm for dealing with such pollutants may be found in^[1] and references cited in.

The following Section will be dedicated to applying such technique to kill microorganisms.

5. Disinfection

As aforesaid, a benefit of UV-founded oxidation techniques remains the concurrent disinfection of the raw water.^[1,26,27] UV disinfection is mostly attributed to the absorption of UVC by deoxyribonucleic acid (DNA) and ribonucleic acid (RNA) which provokes the generation of dimers. Thus, the primary emission of the LPMVL at 254 nm gives rise to demobilizing microbes. Disinfecting performance of VUV light is a function of the work of oxidative species which causes surface oxidation on the cell wall and cell membrane molecules.^[7] Further, hydroxyl radicals can spread into the cell to demobilize enzymes, deteriorate intracellular components, and interfere with protein synthesis.^[28]

Researchers^[29] established dissimilarity in the demobilization of *Xanthomonas* sp. in pure water among UV and VUV irradiation. In pure water, the level of hydroxyl radicals is bigger juxtaposed to

water which holds radical scavengers. The bigger $\bullet\text{OH}$ level conducts to a quicker demobilization below VUV irradiation contrasted to UV irradiation solo.^[1]

In the existence of radical scavengers, for instance, natural organic matter (NOM), the demobilization is mostly attributed to the 254 nm emission of the LPMVL. Researcher^[30] studied the demobilization of coliform bacteria, *Escherichia coli*,^[31-32] and bacteria^[33-35] separated from the sediment of a potable water reservoir.^[36-38] Demobilizing coliform bacteria^[39-42] and *E. coli* was observed to be linear with elevating irradiation period and depicted no distinction among the irradiation at 254 and 185 nm and at 254 nm sole if potable water was utilized as a matrix for the irradiation trials. The separated bacteria from the sediment of a potable water reservoir could not be demobilized entirely because of the existence of spore-forming species such as *Clostridium perfringens*. Spores are indeed more resistant to UV irradiation than the vegetative forms.^[7] The same scientist^[30] established that demobilizing *Bacillus subtilis* spores did not change among VUV irradiation at 254 and 185 nm and UV irradiation at 254 nm. For both wavelengths, the irradiation period requested for total demobilization was much bigger than for coliform bacteria.^[1]

Researchers^[7] explored the demobilization of *B. subtilis* spores at 172 nm irradiation juxtaposed to the irradiation at 222 and 254 nm. The findings depicted that the VUV span was much less efficacious than the tried UV irradiation wavelengths. The spores appeared to be even more resistant towards the demobilization via hydroxyl radicals.^[28] Such results prove that VUV irradiation solo, for example from a Xe-excimer source, stays not appropriate for killing pathogens; however, using LPMVLs with emission in the UV and VUV span remains convenient for integrated disinfection/advanced oxidation methods.^[1] As the disinfection performance of VUV irradiation is much smaller than that of the UVC irradiation (in the existence of radical scavengers), the learning of the UVC injection is satisfactory for anticipating the demobilization.

Introducing ozone which can be produced photochemically within the device improves the disinfection performance of the setup.^[1] As an illustration, bacteria (comprising spores) separated from the sediment of a potable water reservoir can be demobilized totally if ozone was introduced to the UV reactor.^[30] Introducing ozone leads to the demolition of cell membrane and deteriorates the enzymatic activity of UV resistant bacteria.^[43] Furthermore, ozone is an efficacious agent for viruses that are resistant to UV disinfection.^[22] Scientists^[22] developed a UV setup with internal ozone production via VUV irradiation for the synergistic disinfection by UV and ozone.

UV and the integration of the irradiation technologies with ozone remain suitable demobilization processes; however, they do not furnish a disinfectant residual and the regrowth of the bacteria may happen in the distribution system.^[1] Scientist^[30] followed the regrowth possibility following UV and VUV irradiations. The regrowth possibility following VUV irradiation was slightly bigger than after UV irradiation because of the generation of lower molecular weight compounds throughout the photo initiated oxidation of the water matrix (see Section 5.2 in^[1]). Researchers^[44] studied the influence of ozone on the regrowth possibility and detected bigger rates in the total cell count following ozonation; however, not for *E. coli*. To avert

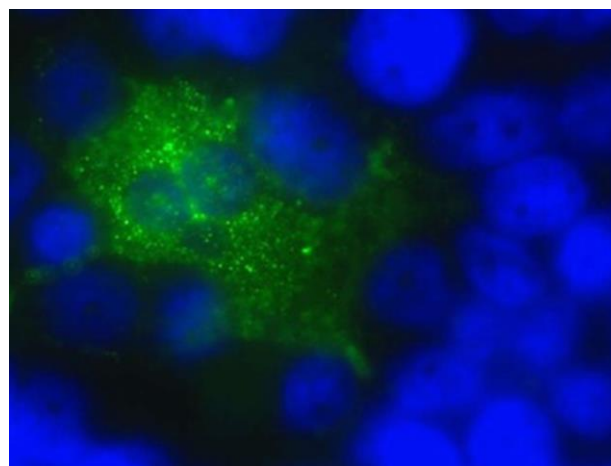


Fig. 3. Immune fluorescence of Hep2G/C3A cells 5 days after infection with Hepatitis E Virus (p6Kernow strain). Cells were stained with Anti-Hepatitis E Virus ORF2,1 Mouse Antibody (MAB8003, Millipore) and Alexa Fluor®488 Goat Anti-Mouse IgG(H + L) Antibody (A11001 Life Technologies).^[46]

the regrowth after disinfection with UV or ozone, implementing an auxiliary disinfectant stays suggested. Inventors^[45] proposed the integration of a UV setup with internal ozone production and a silver cartridge.

6. Designing and implementing VUV irradiation in water treatment

Zoschke et al.^[1] deeply reviewed the reactor design. Therefore, more details about designing vacuum-UV radiation at 185 nm devices may be found in^[1] and references cited in. In a similar fashion, more information about VUV irradiation for treating water, as well as treatment costs, is explained in.^[1]

Hepatitis E Virus (HEV) stays a main reason of waterborne eruptions in regions with poor sanitation. Since secure water supplies are the keystone for avoiding HEV eruptions, information on the effectiveness of disinfection techniques are requested. Guerrero-Latorre et al.^[46] focused on the capability of UV radiation^[47-49] and flocculation-chlorination sachets (FCSs) to decrease HEV in water matrices. The HEV-p6-Kernow strain was replicated in the HepG2/C3A cell line, and the genome number was determined employing qRT-PCR and infectivity using an immune fluorescence assay (Fig. 3). UV irradiation trials employing low-pressure radiation depicted inactivation kinetics for HEV of 99.99% with a UV fluence of 232 J/m² (IC 95%, 195, 02-269,18). Further, the FCSs preparations greatly decreased viral levels in both water matrices.

7. Conclusions

From this work, the following conclusions can be drawn:

1. Improving VUV light sources has revealed novel potentials for the in situ production of hydroxyl radicals and the VUV photolysis of water stays a domain of buoyant investigation juxtaposed to different AOPs.^[1] Several types of research have proved that irradiating water via 185 nm VUV light conducts to the quick decomposition of organic micro-contaminants and a consecutive

degradation of bigger NOM molecules. Further, the UV emission of LPMVLs at 254 nm conducts to concomitant disinfection of the water. Integrating VUV irradiation of water and producing ozone via VUV in the gas phase provides several chances for treating water in the future. Integrating VUV and ozone formed via the identical lamp ameliorates the oxidation and disinfection performance of the device.

2. Nevertheless, as concluded by Zoschke,^[1] presently at most a few functional implementations of VUV irradiation at 185 nm occur. The weak permeation of the VUV light stays the principal practical restriction of VUV irradiation.

3. Practically, VUV irradiation stays not a true stand by to traditional methods in water treatment, like adsorption on activated carbon, or to diverse AOPs because of the imposed restrictions of the VUV technique. Nevertheless, VUV irradiation bids fresh potentials for particular utilization such as the provision of ultrapure water or as a principal treatment method for decentralized setups. Like for many AOPs, the most suitable use in (large-scale) water treatment remains the implementation as a pre-treatment to improve biodecomposition.^[1]

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Conflicts of Interest

The authors declare no conflict of interest.

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