

# Safety assessment of reactors fitted with ultraviolet lamps and the efficacy of these systems for the disinfection of water intended for human consumption

**Report and Guidelines** 

November 2010 (Request 2009-SA-0002)

## Foreword

Based on the proposal of the Expert Committee (CES) on Water, on 8 January 2009 the French Food Safety Agency (AFSSA) issued a formal internal request to develop guidelines for assessing the safety of ultraviolet (UV) radiation lamps and the efficacy of these systems for the treatment of water intended for human consumption (Request no. 2009-SA-0002).

In light of the experience gained since the 1980s on these treatment systems, AFSSA wished to update the recommendations related to low-pressure mercury vapor lamps and propose new ones for medium-pressure mercury vapor lamps that have been introduced more recently.

A Working Group was created for this purpose by a decision of AFSSA's Director General on 9 January 2009.

These guidelines, prepared for the experts and manufacturers concerned, are intended to clarify the procedures for assessing the safety and efficacy of UV reactors and their conditions of use.

A meeting with the professionals responsible for placing UV reactors on the French market was held at the beginning of the Working Group's mandate to gather information on current practices and on potential difficulties encountered during preparation of the authorisation applications. Existing standards in European Union Member States and in third countries for validation of UV reactors used to disinfect water intended for human consumption were taken into account.

The Working Group report was adopted by the CES on Water at the 6 July 2010 meeting.

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TROJAN UV	ABIOTEC
OZONIA	

## **Definitions and glossary**

**Biodosimetry:** method for measuring the dose of UV radiation delivered by a UV reactor by using a dosimeter.

**Challenge microorganism:** microorganism whose sensitivity to UV radiation has been determined under static laboratory conditions.

**Chromophore:** a set of atoms responsible for a molecule's colour. The colour comes from the ability of this assembly of atoms to absorb the energy of photons in a UV-visible spectral range while other wavelengths are transmitted or scattered. In living organisms, chromophores serve to detect light (photoreception) or absorption of light energy (photosynthesis).

**Cofactor (or coenzyme):** the non-protein part of the enzyme. To be distinguished from the apoenzyme, which is the protein part of the enzyme. For example, photolyase is an enzyme composed of a redox-cofactor, FAD, noncovalently bound to the apoenzyme.

Contact time (t): the time during which water and/or microorganisms are exposed to UV radiation.

Dark-repair: DNA repair mechanism by enzymatic reaction (independent of photon energy).

**Deposited dose**: dose actually received by the microorganism, calculated from data generated in laboratory tests.

Dose (D): amount of energy received by a microorganism subjected to radiation of intensity i for time t

$$D = I \times t$$
  
Ws/m<sup>2</sup> or J/m<sup>2</sup> units

**Fluence rate (E'):** radiating power emitted in all directions, passing through an infinitesimally small sphere, of cross-sectional surface area dA, divided by dA. The fluence rate is expressed as W/m<sup>2</sup>, but for convenience,  $\mu$ W/cm<sup>2</sup> or mW/cm<sup>2</sup> is used. This term is equivalent to irradiance in the field of UV reactors.

**Hydraulic residence time (theoretical):** theoretical residence time of water in the reactor, obtained by dividing the volume of the UV reactor by the circulating flow rate.

**Irradiance (E) or flux of radiation:** measured value of the luminous flux divided by the surface area that received the radiation. It is expressed as W/m<sup>2</sup>.

**Minimum required dose**: dose needed to obtain the level of disinfection sought in terms of the reduction desired.

**Molar extinction (or absorption) coefficient (** $\epsilon$ **):** Defined in a given wavelength, it expresses the ability of a molecule to absorb the radiation in question, depending on the medium (solvent) and the temperature. It is expressed as: L.mol<sup>-1</sup>.cm<sup>-1</sup>.

Photoreactivation: a metabolic mechanism of DNA repair induced by photon energy.

Purines: nitrogenous bases forming DNA and RNA molecules (adenine, guanine).

**Pyrimidines:** aromatic heterocyclic nitrogenous bases with the molecular formula  $C_4H_4N_2$ . Pyrimidines are the set of derivatives found in the nitrogenous bases that make up molecules of DNA and RNA (cytosine, thymine, uracil). These bases are particularly affected in photochemical reactions under UV radiation.

**Quantum efficiency (** $\Phi$ **):** the number of molecules photolysed compared to the number of photons absorbed over the same time.

**Solarisation:** the effect of visible and UV radiation (<380 nm) on the envelopes of the lamps making them more or less opaque over time. Doping the quartz with cerium oxides helps to reduce this phenomenon.

**Transmittance (%)**: ability of a medium (water here) to allow the passage of UV radiation. The higher the transmittance, the greater the UV radiation penetrating the water.

**UV or fluence dose (H'):** total radiating energy passing through an infinitesimally small sphere, of cross-sectional surface area dA, divided by dA. The UV dose is the fluence rate (E') multiplied by the irradiation time expressed in seconds. The UV dose is expressed as  $J/m^2$ , but the UV dose is often given as mWs/cm<sup>2</sup> or mJ/cm<sup>2</sup>.

**UV reactor**: device for treating water with UV radiation.

### **Abbreviations**

AFSSA: French Food Safety Agency

MA: Marketing Authorisation

ANSES: French Agency for Food, Environmental and Occupational Health & Safety

CSP:French Public Health Code

WIHC: Water Intended for Human Consumption

ENDWARE: European Network of Drinking WAter REgulators

FAD: Flavine Adenine Dinucleotide

UV:Ultraviolet radiation

LP:Low Pressure

**MP:Medium Pressure** 

CFU:Colony Forming Unit

PFU:Plaque Forming Unit

W/cm: Watt per centimetre of lamp length

## Introduction

Water intended for human consumption is a vital food. As such it must be distributed continuously, in sufficient quantity and be of the best possible quality. Specific purification treatments are evolving as innovations are introduced. These innovations can address health or technical problems, but it is necessary to ensure their efficacy and safety for health in terms of the organoleptic, chemical and/or microbiological qualities of the water.

The first applications of UV irradiation treatment to drinking water for the disinfection of water intended for human consumption (WIHC) date from the early twentieth century, as a result of the development of mercury vapor lamps and quartz sleeves surrounding the lamps, and studies establishing the biocidal effect of these systems (Henry *et al.*, 1910). Currently, these reactors are used in water purification plants, particularly for their action against *Cryptosporidium* and *Giardia*. These microorganisms cause outbreaks of infection in humans through consumption of contaminated water and the conventional biocides used to disinfect WIHC are not effective for inactivating them (WHO, 2009).

The regulatory provisions concerning UV reactors fitted with low-pressure mercury vapor lamps, and used for conventional bactericidal treatment, are specified in a 1987 Circular (Directorate General for Health [DGS], 1987). For claims relating in particular to the inactivation of protozoa belonging to the genera *Cryptosporidium* and *Giardia* and the use of medium-pressure mercury vapor lamps, UV reactors are considered to be innovative systems. According to the French Public Health Code (CSP) (Articles R.1321-1 *et seq*), assessment of the safety and efficacy of innovative treatment products and systems is based on a dossier containing the elements defined in the Ministerial Order of 17 August 2007 as amended (DGS, 2007). Since its creation AFSSA, and now ANSES, has been in charge of scientific and technical assessment of these marketing authorisation (MA) application dossiers.

From its experience acquired with authorisation applications for UV reactors, and taking into account developments in knowledge, claims, technologies and analytical methods, and consistent with the EU context relating to these treatments, ANSES proposes in this report guidelines for assessing UV reactors used for the disinfection of water intended for human consumption.

Confronted with many developments in UV reactors, this report covers only UV radiation products using mercury vapor lamps (low pressure, low output; low pressure, high output and medium pressure) installed in the flow of water to be treated. In addition, the guidelines offered in this report only apply to the production of water intended for human consumption in drinking water treatment plants.

## **1** Overview and description of the systems using UV reactors

The systems currently marketed in France operate either with low-pressure (LP) mercury vapor lamps or medium-pressure (MP) mercury vapor lamps.

#### **1.1.General information about UV radiation**

In the electromagnetic spectrum, UV radiation lies between X-rays and the violet of the visible spectrum, in the range of wavelengths between 100 and 400 nm.

The UV light spectrum is divided into four regions based on the effects on living matter:

- UV-A: 400 to 315 nm, producing skin pigmentation,
- UV-B: 315 to 280 nm, for vitamin D synthesis,
- UV-C: 280 to 200 nm, used for their biocidal power. This is the area considered for water disinfection,
- UV < 200 nm, rays from this part of the UV spectrum induce, for example, the production of ozone from oxygen, and are absorbed by most media.



Figure 1: Distribution of wavelengths in the UV range (from US EPA, 2006)

Atoms have the ability to absorb energy with the promotion of an electron to a higher activated 'excited' energy state. When the electron reverts to its ground state, the loss of excess energy results in the emission of light energy. This energy is quantized. It is equal to:

#### $\Delta E = hv = h c/\lambda$

h = Planck constant

- c = speed of light
- $\lambda$  = wavelength of the radiation emitted by the atom

The value of the wavelength obtained depends on the energy difference between the activated state and the ground state.

The activation of mercury atoms is the most widespread technology for the generation of UV radiation used in water treatment. Mercury atoms excited to higher energy levels will generate a line spectrum (meaning the emission of luminous radiation at different discrete wavelengths, of varying intensity) when they return to the ground state. Mercury is the most volatile metal for which activation can be achieved in the gas phase at temperatures consistent with the materials used to manufacture the lamps. At ambient temperature, before the lamps are switched on, the mercury contained in them is essentially in a liquid state and non-conductive of the electric discharge. To initiate the discharge, the

lamps contain a gas, called a "buffer", that is more easily excitable. The most commonly used buffer gas is argon. Neon and helium can also be used. Generally, this gas is in excess of the gaseous mercury. Its role is to facilitate discharge initiation and promote excitation of the mercury.

#### 1.2.Lamps used for treatment with UV radiation

UV radiation is generated by the arc of light from electric discharge lamps. For water disinfection, lamps that have been fitted with plasma emitters are used. The emission of photons by an atom is a reversible phenomenon. Thus, a photon generated within plasma can be absorbed reversibly by a mercury atom.



#### Figure 2: Diagrams of low and medium-pressure mercury vapor lamps (US EPA, 2006)

Both types of lamps (LP - low-pressure and MP - medium-pressure) emit in the UV-C range with a quasi-monochromatic spectrum for low pressures at 253.7 nm, and polychromatic spectrum for medium pressures for which the wavelengths range from UV-C (200 to 290 nm) to UV-A (315 to 400 nm) but also up to visible radiation. Currently, new types of lamps such as those described in Annex E are on the market but are not yet authorised in treatment plants producing water intended for human consumption.

#### 1.2.1.Mercury vapor lamps

#### 1.2.1.1.Low-pressure lamps (LP)

**Low-pressure lamps** operate with mercury vapor pressure ranging from about 100 to 1000 Pa; a buffer gas supplements the mercury vapor. The buffer gas exceeds the mercury vapor by 10 to 100 times. Liquid mercury is still found at the nominal operating equilibrium temperature. The partial mercury vapor pressure inside the lamp is approximately 1 Pa.

The lamps are powered by a low-frequency alternating current (50 Hz). In the event of a voltage drop, the emission is practically extinct.

These lamps have the benefit of quasi-monochromatic emission with an emission line at 253.7 nm. There is a second line at 184.9 nm but it is generally eliminated by the use of optical lenses, quartz doped with titanium dioxide or air separating the lamp from the protective sleeve. Emissions at wavelengths above 300 nm are also found but may be disregarded in water treatment. The Austrian ÖNORM M.5973-1 Standard specifies that emission at 253.7 nm must be at least 85% of the total UV-C (200-280 nm) intensity.

The emission line at 253.7 nm (Figure 3) corresponds to a wavelength near the maximum absorption of DNA and RNA. This results in greater efficacy of these LP lamps since most energy is emitted at a biocidal wavelength. However, the power of these LP lamps is limited to several hundred biocide Watts. They also have the disadvantage of being very long at that power level: up to 1 or even 1.5 metres in length.



Figure 3: Emission spectrum of a low-pressure mercury vapor lamp (DVGW W294-1, 2006) (emission line at 184.9 nm does not appear on the spectrum).

Two parameters affect the performance of low-pressure lamps:

- the temperature of the lamp envelope that acts directly on the mercury vapor pressure along the inner envelope of the lamp. If it is too low, there is a drop in the emission yield; if it is too high, the mercury vapor pressure is increased and self-absorption increases, which reduces the emission yield. For this reason, emitting lamps are installed in a quartz sleeve inside which air flows to moderate the water cooling effect.
- the aging of the lamps: over the first 100 to 200 hours of operation, a drop in emission yield is observed. The cause of aging is the solarisation of the lamp envelope material and the

formation of oxide deposits on the electrodes. Each ignition/extinction operation corresponds to aging equivalent to the first hour of operation under nominal conditions.

Low-pressure lamps require a temperature of approximately 40°C and consume 0.5 to 0.6 W/cm (per centimetre of lamp length) of electrical power.

Low-pressure high-output lamps operate at approximately 90°C. The electrical power consumed ranges from 2 to 3 W/cm for an emission from 0.3 W/cm to 0.5 W/cm. Their energy output is an advantage but they take longer to heat up in order to reach peak performance.

#### 1.2.1.2.Medium-pressure lamps (MP)

**Medium-pressure lamps** operate with mercury vapor pressure of approximately 10 kPa, which enables this type of lamp to reach electrical power levels 100 times higher than low-pressure lamps, i.e. several tens of W/cm. However, the counterpart of this gain, in terms of electrical power, is the occurrence of a quasi-continuous emission spectrum between 200 and 300 nm, and emission mostly at wavelengths above 300 nm (for the other mercury emission lines between 300 and 600 nm), which reduces the biocidal yield by 10 to 15%. They may have titanium-doped quartz sleeves that cut all wavelengths below 230 nm, thus limiting reactions with other substances in water that can form undesirable compounds (see § 2.3).



Wavelenght (nm)

Figure 4: Emission spectrum of a medium-pressure mercury vapor lamp (from ÖNORM, 2003)

Under normal operating conditions, no liquid-phase mercury remains in the lamp.

Medium-pressure mercury vapor lamps operate in a voltage range from 5 to 30 V/cm. The UV energy emitted is proportionate to the power supply voltage, which also determines the average electrical output of the lamps' power supply (80 to 250 W/cm). By increasing the electrical power, emission bands are widened. This phenomenon is a constraint that will be dealt with in the emission spectrum section.

The coldest part of a medium-pressure mercury vapor lamp reaches temperatures of approximately 400°C. Contact between the envelope of the lamp and the water is prohibited. These lamps must be inserted into a ventilated quartz sleeve to lower the temperature of the envelope in contact with the water to be treated.

The generally accepted lifetime for medium-pressure lamps is from 8,000 to 10,000 hours. Aging results in a change in the emission spectrum.

Emission from medium-pressure mercury vapor lamps is polychromatic with numerous emission lines in the UV range as well as in the visible and infrared regions.

The ÖNORM M 5873-2 Standard stipulates that lamps used for water disinfection must not emit, at wavelengths below 240 nm, energy higher than 3% of that emitted at wavelengths between 240 and 400 nm. The DVGW W294-1 Standard specifies that the radiant power in water, in the wavelength range below 240 nm, must be less than 5% of the total radiant power between 240 and 290 nm.

This is because a large amount of energy from wavelengths below 240 nm can lead to secondary reactions (see § 2.3.). The lamp's emission spectrum should be measured at low lamp power. Unlike low-pressure lamps, the power emitted at the 253.7 nm wavelength should not exceed 85% of the power emitted over the entire UV-C range.

Table I summarises the characteristics of different lamps.

Parameters	Low-pressure lamp	High-pressure high- intensity lamp	Medium-pressure lamp
Length (m)	0.2 to 1.5	0.1 to 1.5	0.2 to 1
Diameter (cm)	1.5 to 3		1.5 to 4.5
Light	Monochromatic at 254 nm	Monochromatic at 254 nm	Polychromatic including 200 – 300 nm
Mercury pressure (Pa)	Approximately 0.93	0.18 to 1.6	40,000
Temperature (°C)	Approximately 40	60 – 100	600 – 900
UV dose applied (W/cm)	0.2	0.5 to 3.5	5 to 30
Electrical power consumed (W/cm)	0.5	1.5 to 10	50 to 250
Electrical to germicidal UV conversion efficiency (%)	35 to 38	30 to 35	10 to 20
Relative number of lamps needed for a given dose	High	Intermediate	Low
Lifetime (hours)	8000 – 10,000	8000 - 12,000	4000 – 8000

Table I: Characteristics of mercury vapor lamps (US EPA, 2006)

#### 1.2.2.Quartz sleeves

The lamps are housed within a quartz sleeve that is approximately 2.5 cm in diameter for low-pressure lamps and from 5 to 10 cm for medium-pressure lamps. The distance between the lamp and the sleeve is 1 cm.

For medium-pressure lamps, the sleeves are made of doped quartz (titanium dioxide for example) to absorb radiation at wavelengths below the limit used to reduce adverse photochemical reactions. Some manufacturers use protective films, which also absorb wavelengths.



Figure 5: Transmission spectrum of doped quartz and non-doped quartz (US EPA, 2006)

#### Cleaning the sleeves

These sleeves attract deposits (calcium carbonate, metal oxides, etc.) when in contact with water and must be kept clean in order to ensure proper radiation transmission. Two cleaning procedures are commonly used:

- mechanical cleaning that can be done during water treatment cycles with a Teflon® scraper ring that runs the length of the sleeve regularly;
- chemical cleaning that is implemented outside treatment cycles using an acid solution (citric acid, phosphoric acid) that is pumped through the shut down reactor, after which a flushing sequence is activated.

#### 1.2.3.Ballasts

Ballasts ensure the power supply to the lamps, starting of the lamps, and control of the electric power transmitted. There are three types of ballast: inductive, capacitive and electronic.

In the case of capacitive ballasts, the intensity of the current flowing through the lamp does not vary with the voltage applied. With inductive ballasts, the current flowing through the lamp depends on inductance, the voltage applied and the lamp's properties. Inductive ballasts are mostly used for medium-pressure lamps, because of their durability and stability.

Electronic ballasts contain semiconductors.

#### **1.3. Reactors**

#### 1.3.1.Components of a reactor

Electrical energy, provided by one ballast, excites mercury vapors to an energy level sufficient for emitting UV radiation. The lamp is placed in a casing and water flows in a thin layer between this casing and the quartz sleeve, because the water quickly absorbs the UV radiation energy. The casing can contain one or more lamps depending on the flow to be treated and the energy to be generated.

Water absorbs UV radiation more intensely than visible light, and UV rays weaken in proportion to their distance from the source. Therefore the transmitters must be placed very close to each other.

An exponential decrease in the penetration of UV radiation results from a linear increase in the thickness of the layer and an increase in the UV absorbance of the water.

#### **1.3.2.Operating hydraulics of the reactors**

The first reactors used were designed for low flow rates (5  $m^3/h$ ). In this case, the configuration is that of a tubular reactor with a lamp surrounded by a protective sleeve installed at the centre. The water flows between the protective sleeve and the reactor's inner casing. According to the Beer-Lambert law, the intensity decreases exponentially between the envelope of the protective sleeve and the reactor casing.

Figure 6 shows a UV radiation disinfection equipment.



Figure 6: Example of a UV radiation disinfection equipment (US EPA, 2006)

The reactors used for the production of drinking water are closed reactors.

Depending on the technology, the lamps can be positioned axial or perpendicular to the flow as shown in Figures 7 and 8.

In some reactors, they are placed apart from the flow of water and the rays are reflected onto the hydraulic path.



Figure 7: Axial position of the reactor relative to the flow (US EPA, 2006)



Figure 8: Perpendicular position of the reactor relative to the flow (US EPA, 2006)

#### 1.3.3.Hydraulic modelling

The dose distribution in a UV reactor depends on many variables that are often interdependent: reactor shape, number, spacing and power of the lamps, characteristics of the lamp sleeves, layout of the deflectors, transmittance and velocity of the water (see §1 of Annex A).

That is why design and development of the UV reactors by the manufacturers generally go through a modelling step that combines two sub-models, applied in succession:

- a distribution model of the rate of irradiation (fluence) in the reactor, in order to describe the spatial variation of the UV energy in the reactor. In this step, the reactor is considered to be thoroughly agitated;
- a hydrodynamic model to take into account turbulence and fluid flow phenomena within the reactor, this considers the flow or velocity gradient of the microorganisms assimilating the particles.

It should be noted that recent developments make it possible to operate these two models simultaneously.

#### **1.4.Control systems**

UV intensity sensors (or radiometers) are photosensitive detectors used to determine the UV dose delivered, by measuring the intensity at different points of the reactor. Based on their position, the radiometers can also respond to changes in UV absorbance in the water treated.

#### 1.4.1. Radiometers

Radiometers are UV energy sensors made up of optical components, a photodetector, an amplifier (to convert the photodetector's electrical signal into an irradiance value) and an electrical connection.



Figure 9: Examples of radiometers (a. fitted to an LP reactor; b. fitted to an MP reactor) (US EPA, 2006)

An ideal radiometer should provide a linear response in the working field, independent of the temperature, and should be stable over time, and insensitive to background noise and bias (Figure 10a). In addition, it should have an angular response as close as possible to the cosine. Figure 10b shows two radiometer angular responses: radiometer 1 is close to ideal; radiometer 2 does not measure all of the incident light.



Figure 10: Ideal criteria for radiometers. (a. Linearity criteria / b. Angular response) (US EPA, 2006)

The set of parameters to be taken into account for assessing the performance of radiometers is detailed in Annex B.

The monitoring radiometer fitted to UV reactors must be calibrated at least once a month using a reference radiometer (recommendation of the US EPA, 2006). In addition, according to the same source, reactors fitted with medium-pressure lamps must have one radiometer per lamp and those fitted with low-pressure lamps must have one per row of lamps.

#### 1.4.2. Reference radiometers

Reference radiometers are measuring instruments independent of those used to monitor the efficacy of UV disinfection devices. They must be calibrated in authorised laboratories with a control radiometer for radiation of 253.7 nm. The calibration uncertainty must be less than or equal to 6%. After 100 hours of operation and at least every two years, the reference radiometers must undergo standardisation again and re-calibration.

The devices' radiometers must be calibrated such that their measurement value corresponds to that of the reference radiometer. After 10,000 operating hours and at least every two years, they must again be calibrated and standardised. If the measurement value of the device's radiometer is greater than 5% of the reference radiometer value, calibration is necessary.

The measurement range and the UV reactor radiometer display must be adapted to the radiant power to be monitored. The radiometer measurement heads must be labelled so that their type, manufacturer, serial number and properties are clearly recognisable in order to determine whether the radiometer's type and measurement range are suitable for the UV reactor concerned.

#### 1.4.3. Monitoring the power consumption of the lamp

Continuous measurement of the reactor's power consumption indicates whether the lamp is operating correctly or displays abnormal aging. There is a correlation between the power supply (U in Volts) or wattage output (P) and the UV energy emitted (P=IU in Watts).



Figure 11: Correlation between the power supply or wattage output and the UV energy emitted (Masschelein W. J., 2000)

(I = total UV energy between 240 and 380 nm in relative units; e = electric power supply). The precise correlation between the emitted energy I, and the electric power supply W (e) also depends on the ballast and the transformer, but it is important to note that the correlation is almost linear for a given design and construction.

# 2. Benefits and risks associated with the use of the systems implementing UV reactors

One of the disinfection stages in the process of treating water intended for human consumption (WIHC) may include UV reactors. They facilitate local disinfection without inducing a residual effect as can take place with some chemical reagents approved for the same purpose. Their inactivation efficacy has been shown on protozoa of interest such as *Cryptosporidium* and *Giardia*, against which conventional disinfectants do not have the required efficacy (WHO, 2009). These technologies are therefore of interest in safety to health of WIHC.

#### 2.1.Mechanisms of action

In a simple photochemical reaction, a compound A is converted into one or more compounds, under the action of mono- or polychromatic irradiation.

The photolysis quantum yield  $\Phi_{\lambda}$  of compound A is defined at the irradiation wavelength  $\lambda$  as the number of degraded molecules per absorbed photon.

Thus compound A can be photolysed at wavelength  $\lambda$  by direct means only:

- if A absorbs the UV radiation, which means that its molar absorption coefficient  $\epsilon_{\lambda}$  at wavelength  $\lambda$  is not negligible;
- if the quantum yield  $\Phi_{\lambda}$  is sufficiently high.

The efficacy of the photochemical reaction will be directly proportional to the product  $(\epsilon_{\lambda} \times \Phi_{\lambda})$ .

The whole spectrum of UV radiation can initiate photolysis reactions with the mineral elements or organic compounds found in water, but the formation of secondary compounds is particularly evident for the highest energy UV radiation at wavelengths below 230 nm.

With low-pressure mercury vapor lamps, secondary reactions occur at irradiation doses approximately equal to or higher than 1000 J/m<sup>2</sup>; this is much higher than those commonly used for disinfection (250 to 400 J/m<sup>2</sup>). Indeed, low-pressure mercury vapor lamps emit quasi-monochromatic radiation at 253.7 nm, which substantially limits the risks of formation of undesirable compounds.

Medium-pressure mercury vapor lamps emit polychromatic radiation at wavelengths ranging from 200 nm to 800 nm (but only 10 to 15% of the radiation power is emitted at wavelengths of between 200 and 280 nm). In practice, these medium-pressure lamps, used for disinfection of the public water supply, emit in the range of 230-240 nm to 800 nm wavelengths because they are all fitted with devices for stopping UV radiation at wavelengths below 230-240 nm. This is to limit the risk of secondary reactions.

#### 2.1.1.Action of UV-B and UV-C radiation on microorganisms

The biocidal effect of UV radiation (UV-B and UV-C: range 200-315 nm) is mainly due to the fact that they are absorbed by DNA and RNA, molecules that support replicative and metabolic functions. DNA and RNA consist of a sequence of nucleotides that includes purine bases (adenine, guanine) and pyrimidine bases (cytosine, thymine or uracil). The absorption spectra of the four DNA nucleotides (Figure 12) show maximum absorption at approximately 260 nm.



Figure 12: Absorption spectra of nitrogenous bases (Kowalski, 2009)

This absorption induces degradation reactions particularly involving pyrimidine bases of DNA (thymine and cytosine) and RNA (uracil and cytosine). Dimerisation reactions are then induced at sites containing two adjacent pyrimidines. Several types of photoproducts are then formed: cyclobutane-type dimers (70 to 80% of photoproducts) and pyrimidine (6-4) pyrimidone adducts (20 to 30%). The latter can then be converted photochemically by UV-As into Dewar valence isomers (Figure 13). UV-Bs can thus generate three types of photoproducts for each of the four pyrimidine doublets, which therefore generates 12 lesions in the genome of a DNA strand (Banyasz *et al.*, 2009).



Figure 13: Chemical structure of dimeric pyrimidine photoproducts induced by UV-B and UV-C radiation in DNA.

The figure shows only thymine – thymine (TT) lesions but equivalent compounds can form between thymine and cytosine or between two cytosines (from Banyasz *et al.*, 2009).

During the formation of cyclobutane pyrimidine dimers (CPDs), the hydrogen bonds that were present between the two adjacent pyrimidines (the example of thymines is shown in Figure 13), are weakened by the loss of aromaticity. In addition, the cyclobutane bonds formed disrupt the structure of DNA, thus

creating a distortion in the DNA helix. This damage blocks replication and transcription, which results in cytotoxic and mutagenic effects, inducing death in the irradiated microorganisms (Cheung *et al.*, 1999).

#### 2.1.2. Action of UV-A radiation on microorganisms

The effect of UVA rays (315 - 400 nm) is related to oxidation processes. DNA does not absorb them but it can excite endogenous chromophores that are responsible for oxidative reactions attributed to the formation of active species, especially singlet oxygen (Cadet *et al.*, 2009). These radical species induce lethal or sublethal effects in cells (Oppezzo and Pizarro, 2001).

#### 2.2.Efficacy

#### 2.2.1.Kinetics of inactivation

During disinfection of WIHC, and depending on the doses used, the claimed efficacy for disinfection will be different and dependent on the type of target microorganism. In all cases, the recommendations for use must be specified in order to achieve the efficacy sought.

The constituents of the water can affect the performance of UV disinfection. The parameters most sensitive to UV absorbance are the particle content of the water, the concentration of elements (iron, for example) that can cause fouling of the reactor, and algae.

The transmittance of the water is very important. If it decreases, the energy within the reactor also decreases, reducing the dose delivered. Thus, as for all other disinfectants, the water must first be thoroughly clarified.

While chemical disinfection uses the notion of **C.t** (biocide concentration and contact time), disinfection by UV irradiation is based on the concept of dose. The dose may be defined by the following equation:

#### Dose = P.t

#### **P** is the biocidal power delivered

#### t is the contact time

The main difference between the concepts of P.t and C.t, is the absence of residue in the reactor outlet with UV, while there is a residual concentration of biocide with a chemical disinfectant.

At the same time, the use of these two concepts depends on a number of parameters.

- The product P.t depends on the physico-chemical quality of the water, the emission spectrum of the lamp, the reduction required, and the initial concentration of microorganisms.
- The product C.t depends on the physico-chemical quality of the water, the reduction required, and the initial concentration of microorganisms. It also depends on the water temperature and pH, whereas inactivation with UV radiation is independent of these two parameters.

The required UV radiation dose is expressed in Joules per square metre  $(J/m^2)$ . It corresponds to the product of the energy received  $(W/m^2)$  by the irradiation time t (seconds). This time depends on the flow of water to be treated and the size of the reactor.

Exposure dose:

$$D = (P t / S)e^{-kx} (in J/m^2)$$

where:

P: biocidal power of the UV radiation source (in W),
S: UV radiation-emitting surface (in m<sup>2</sup>),
t: exposure time of volume element (in s),
k: UV radiation absorption coefficient of the water to be treated (in m<sup>-1</sup>), this coefficient varies from 2 to 10 m<sup>-1</sup> (0.02 to 0.1 cm<sup>-1</sup>) for water for consumption,
x: thickness of the water layer (in m).

The resistance of microorganisms to UV radiation can vary significantly, from a few  $J/cm^2$  to several tens of  $J/cm^2$ .

#### 2.2.2.Illustration of efficacy

The data described in the literature are derived from laboratory strains that have different sensitivities to those of wild strains from the environment. Without standardised protocols for the preparation and counting of microorganisms, the results can vary between studies. However, inactivation testing is conducted under laboratory conditions with a collimated beam apparatus, usually fitted with a low-pressure lamp. The exposure time associated with the energy measured with a standardised radiometer determines the UV radiation dose expressed in  $J/m^2$ .

#### 2.2.2.1.Bacteria

The sensitivity of bacteria to UV radiation is outlined in Table II.

Doses of UV radiation of between 10 and 100  $J/m^2$  enable inactivation of at least 4-log of pathogenic bacteria and bacterial indicators of faecal contamination, that are thus rendered non-culturable. *Helicobacter pylori* have similar sensitivity since a dose of 80  $J/m^2$  is associated with a reduction of more than 4-log (Hayes *et al.*, 2006).

Bacteria of hydrotelluric origin have sensitivities to UV radiation that vary widely by genera and species. The number of *Acinetobacter baumanii* is reduced by 4-log after exposure to a dose of 48 J/m<sup>2</sup> (Templeton *et al.*, 2009). The amount of *Legionella pneumophila* is reduced by 4-log with a dose of 64 and 94 J/m<sup>2</sup> (Wilson *et al.*, 1992; Oguma *et al.*, 2004). Among mycobacteria, the number of *Mycobacterium avium* is reduced by 4-log with doses of approximately 200 J/m<sup>2</sup> (Hayes *et al.*, 2008; Shin *et al.*, 2008) while the species *M. terrae* requires a dose of 100 J/m<sup>2</sup> for a 2-log reduction only (Bohrerova and Linden, 2006) and the species *M. fortuitum* a dose of at least 500 J/m<sup>2</sup> for a 3-log reduction (Lee, 2009).

Species likely to be associated with bioterrorism, such as *Brucella suis*, *B. melitensis*, *Burkholderia mallei*, *B. pseudomallei*, *Francisella tularensis* and *Yersinia pestis* require doses of UV radiation of at least 120 J/m<sup>2</sup> to achieve 4-log reductions (Rose and O'Connell, 2009). They exhibit the same sensitivity to UV radiation as do waterborne bacteria.

Bacteria in spore form, in turn, are more resistant to UV radiation than those in vegetative form. Thus, doses above 400 J/m<sup>2</sup> are required to reduce *Bacillus anthracis* spore suspensions by 2-log (Rose and O'Connell, 2009) and doses of approximately 800 J/m<sup>2</sup> are necessary to destroy 4-log of *B. Subtilis* spores, the test germ used in the validation tests. DNA is saturated with proteins during sporulation. These bonds inhibit the formation of pyrimidine dimers and promote the formation of 5-thyminyl-5-6-dihydrothymine. Photorepair restores thymines during cell multiplication.

Bacterium	Range of UV radiation doses tested, in J/m <sup>2</sup>	Type of lamp	Maximum log reduction
Salmonella typhi	20-100	LP	5.6
Campylobacter jejuni	5-60 LP		5.3
Yersinia enterocolitica	6-50	LP	5.0
Shigella dysenteriae	10-50	LP	5.9
Shigella sonnei	30-80	LP	4.7
Vibrio cholerae	6-40	LP	5.8
Legionella pneumophila	10-120	LP	4.4
Legionella pneumophila	5-30	LP	3.0
Escherichia coli 0:157	10-70	LP	5.5
Escherichia coli	10-150	LP	6.0
Escherichia coli	15-90	MP	5.2
Streptococcus faecalis	25-160	LP	4.6
Bacillus subtilis (spores)	50-780	LP	4.0
Clostridium perfringens (spores)	480-640	MP	3.0

Table II: Efficacy of UV radiation on the reduction in the number of culturable bacteria (adapted from Hijnen et al., 2006)

#### 2.2.2.2.Viruses and bactieriophages

#### - Animal viruses

Sensitivity of viruses to UV is shown in Table III.

RNA viruses such as enteroviruses, hepatitis A virus, rotavirus and calicivirus are reduced by 4-log with doses lower than or equal to  $400 \text{ J/m}^2$ . There is uncertainty about the norovirus, which cannot be cultured with current methods.

Adenoviruses show greater resistance to UV radiation although with differences depending on the type of lamp used. Thus, a dose of 400 J/m<sup>2</sup> emitted by a low-pressure lamp can only cause a 1-log reduction in infectivity of serotypes 2, 5 and 41. Serotype 41, associated with episodes of diarrhoea, proves more resistant than the other serotypes to pulmonary tropism. While a dose of 1200 J/m<sup>2</sup> results in inactivation of 3-log of serotypes 2 and 5, a 1-log reduction is obtained for serotype 41 (Baxter *et al.*, 2007). To minimise the difficulties associated with *in vitro* cell culture of some adenoviruses, the measurement of inactivation by integrated cell culture – quantitative polymerase chain reaction (ICC-qPCR) has shown a 4-log reduction in serotype 4 (Gerrity *et al.*, 2008).

For a given dose, medium-pressure lamps ensure greater inactivation than low-pressure lamps. A 4-log reduction in serotype 40 is obtained with a 600 J/m<sup>2</sup> dose (Linden *et al.*, 2007).

The lower sensitivity of adenoviruses to UV radiation is related to the ability of the host cells to repair the damaged viral DNA. This form of repair is not found in RNA viruses. Low- and medium-pressure lamps are equally effective at damaging viral DNA but medium-pressure lamps impair other viral targets that are not repaired by the host cells (Eischeid *et al.*, 2009).

Virus	Range of UV radiation doses tested, in J/m <sup>2</sup>	Type of lamp	Maximum inactivation in log
Poliovirus type 1	50-500	LP	5.4
Adenovirus serotypes 2, 15, 40, 41	80-3060	LP	6.4
Adenovirus serotype 40	80-1840	LP	3.0
Adenovirus serotypes 2, 41	300-900	MP	4.3
Rotavirus SA-11	50-500	LP	4.1
Rotavirus SA-11	50-300	MP	4.6
Feline, canine calicivirus	40-490	LP	5.5
Bovine calicivirus	40-330	LP	5.7
Bovine calicivirus	20-150	MP	5.9
Hepatitis A	50-280	LP	5.4
Coxsackie virus B5	50-400	LP	4.8

Table III: Viral inactivation (adapted from Hijnen et al., 2006)

#### - Bacteriophages

Sensitivity of bacteriophages to UV is shown in Table IV.

Bacteriophages are recommended for performing UV radiation efficacy tests because they are safe, and easy to handle in the laboratory. The RNA MS2 phage used in standardised validation protocols is one of the most resistant to UV radiation.

Bacteriophage	Range of UV radiation doses tested, in J/m <sup>2</sup>	Type of lamp	Maximum inactivation in log
MS2	50-1390	LP	4.9
MS2	120-460	MP	5.3
φX174	20-120	LP	4.0
PRD1	90-350	LP	3.8
B40-8	10-390	LP	5.6
Т7	50-200	LP	4.6
Qβ	100-500	LP	4.2

Table IV: Inactivation of bacteriophages by UV radiation (adapted from Hijnen et al., 2006)

#### 2.2.2.3.Protozoa

Sensitivity of protozoa to UV radiation is presented in Table V.

The efficacy of UV radiation related to inactivation of *Cryptosporidium* oocysts and *Giardia* cysts has been demonstrated by testing infectivity (Clancy *et al.*, 1998; Craik *et al.*, 2000). The *in vitro* excystation assays used previously underestimated the inactivation performance of UV radiation.

The meta-analysis by Qian *et al.*, 2004, based on a statistical approach using results from 14 publications, indicates that to obtain a reduction of at least 3-log at a 5% risk level, a dose of 80 to  $140 \text{ J/m}^2$  is required for *Cryptosporidium sp.* oocysts and a dose of 120 to 200 J/m<sup>2</sup> for *Giardia sp.* cysts.

The WHO, in its summary paper on managing risk related to *Cryptosporidium* in WIHC, selected a dose of 90 J/m<sup>2</sup> for a 3-log reduction in *Cryptosporidium* cysts.

Low and medium-pressure lamps have similar inactivation potential against *Cryptosporidium* oocysts (Craik *et al.*, 2001). The same applies to *G. lamblia* cysts (Shin *et al.*, 2009).

Oocysts of *C. hominis*, predominantly found in humans, have similar sensitivity to that of *C. Parvum* oocysts, a bovine strain used in most studies (Johnson *et al.*, 2005). *G. Lamblia* cysts, pathogenic for humans, also show a sensitivity of the same order of magnitude as *G. muris* cysts (Mofidi *et al.*, 2002); however, differences have been identified in other studies (Linden *et al.*, 2002; Craik *et al.*, 2000).

A 4-log reduction in suspensions of *Toxoplasma gondii* oocysts was obtained for a dose of 400 J/m<sup>2</sup> (Dumètre *et al.*, 2008). This performance was not reported by Wainwright *et al.* (2007), probably because of differences in the protocols.

Protozoan	Range of UV radiation doses tested, in J/m <sup>2</sup>	Type of lamp	Maximum inactivation in log
Cryptosporidium parvum	5-61	MP	3.0
Cryptosporidium parvum	9-131	LP	3.0
Giardia muris	15-110	MP	2.4
Giardia lamblia	0,5-15	LP	2.5
Acanthamoeba spp.	430-1720	LP	4.5

Table V: Inactivation of protozoa (adapted from Hijnen et al., 2006)

#### 2.3.Adverse effects

#### 2.3.1.Photolysis reactions

The use of UV radiation for the disinfection of water implies that it has a low absorption spectrum at the wavelengths emitted by the lamps used for disinfection.

All chemical compounds, in solution or suspension, found in water, and absorbing UV wavelengths, can interfere with the efficacy of disinfection by this system. This is the case with suspended solids, turbidity, organic matter of natural origin, some ions (nitrate, nitrite), and some mineral elements (iron) and organic compounds that can cause photolysis reactions and generate undesirable by-products. UV irradiation of water containing a residual oxidant (chlorine, chlorine dioxide, ozone, etc.) results in secondary photochemical reactions that may yield undesirable compounds.

#### 2.3.1.1. Photolysis of nitrate and nitrite ions

Nitrate and nitrite ions are characterised by strong absorption of UV radiation at wavelengths below 230 nm (Figure 14), (Mack and Bolton, 1999).



Figure 14: Absorption spectra of UV/Visible radiation of nitrate and nitrite ions (Mack and Bolton, 1999)

As a result, nitrite and nitrate ions:

- strongly absorb UV radiation at the wavelengths emitted by medium-pressure mercury vapor lamps, especially below 230 nm (for example, 50 mg/L of a nitrate solution produces an absorbance of approximately 1.5/cm at 230 nm);
- absorb only a little UV radiation at 253.7 nm, the emission wavelength of the great majority of low-pressure mercury vapor lamps ( $\epsilon_{253.7}$  nm < 15 M<sup>-1</sup>.cm<sup>-1</sup> for NO<sub>2</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup>).

Nitrate ion photolysis (Golstein and Rabani, 2007) results in the formation of NO<sub>2</sub><sup>-</sup> and oxygen (O<sub>2</sub>) which are the end products of a set of reactions: the first step is photo-isomerisation of the nitrate ion NO<sub>3</sub><sup>-</sup> with the formation of the peroxynitrite ion (ONOO<sup>-</sup>/ONOOH), and dissociation of the ion NO<sub>3</sub><sup>-</sup> with the formation of radicals NO<sub>2</sub>° and OH°. Then a complex reaction mechanism occurs, involving unstable species such as nitrogen oxides (NO°, N<sub>2</sub>O<sub>3</sub>, N<sub>2</sub>O<sub>4</sub>) and hydroperoxyl radicals (HO<sub>2</sub>°/O<sub>2</sub>°<sup>-</sup>), which react to produce nitrite ions, nitrate ions and oxygen.

Photolysis of nitrite ions found initially in water or of those formed during photolysis of nitrate ions also results in the formation of nitrogen oxide (NO°) and hydroxyl radicals (HO°) (Mack and Bolton, 1999). They then initiate reactions similar to those occurring during the photolysis of nitrate ions.

Overall, nitrate and nitrite ion photolysis, in the absence of organic compounds and at wavelengths below 230 nm, cause the formation of unstable compounds in aqueous media such as nitrite, nitrogen oxide and hydroxyl radicals, which recombine to produce the original compounds. Reactions are possible between these radicals and organic compounds (when present), but are unlikely at the doses of UV radiation recommended for disinfection.

#### 2.3.1.2. Photolysis of other inorganic compounds

Some inorganic compounds which are strongly UV-absorbent hinder the disinfectant action of UV radiation, but few data are available in the scientific literature concerning the potential formation of undesirable by-products.

In particular, ferrous ion is known to absorb strongly in the UV range and thus to influence the efficacy of UV irradiation. Ferrous ions have a large number of absorption bands. At the wavelengths used for disinfecting WIHC, even at levels below the reference grade, the oxidation of ferrous oxide into ferric iron, which precipitates as  $Fe(OH)_3$ , and that of manganese into manganese oxide, takes place. These oxides and hydroxides are deposited not only on the sleeves of the lamps, thereby greatly reducing the efficacy of disinfection, but also on the radiometer. For this reason, cleaning and regular maintenance of these surfaces is essential.

#### 2.3.1.3. Photolysis of organic compounds

Organic compounds in water to be treated may undergo photolysis in the UV disinfection stage. This photolysis is only possible if the compound considered has chemical groups capable of absorbing photons (chromophores) emitted by radiation from the UV lamp(s). In the range of UV radiation used in disinfection, this initially means compounds with aromatic groups or double or triple bonds, with a strong conjugation character, which can undergo photolysis (Table VI). Conversely, aliphatic compounds will absorb only very little UV radiation. Kim and Tanaka (2009), for example, have shown that substances containing amide groups of the type R-CON-R<sub>2</sub>, were not very photosensitive, even if they contained an aromatic group (Table VII). Furthermore, their study also revealed that certain drug residues have considerable sensitivity with respect to UV radiation (for example, ketoprofen, Table VII).

λ (nm)	ε (in l.mol <sup>-1</sup> cm <sup>-1</sup> )
220	800
280	12
350	11
240	120
300	100
270	12
271	18.6
218.5	1120
210	1500
	λ (nm)         220         280         350         240         300         270         271         218.5         210

Table VI: Examples of chromophore groups in the 220-350 nm wavelength range (from Kowalski,2009)

The presence of chromophore groups is not sufficient to interpret the photosensitivity of compounds. The structure of the compounds will determine the values of two important parameters for direct phototransformation reactions:

- the molar extinction coefficient (ε) of the compound considered, at the wavelengths of irradiation emitted by the lamp. The higher this coefficient, the more the compound is likely to absorb the UV radiation emitted, and therefore the more likely it is to form by-products;
- photolysis quantum yield ( $\Phi$ ) of the compound considered. Quantum yields are generally independent of the irradiation wavelength (Verhoeven, 1996). However, compounds whose chromophore structure is derived from phenol, including oestrogen-like compounds such as 17- $\alpha$ -ethinylœstradiol, have quantum yields of photoionisation that increase when the wavelength of irradiation decreases (Grabner *et al.*, 1977; Jin *et al.*, 1995).

In addition, the simultaneous presence of radical precursors (organic matter, nitrate ions, etc.) may result in indirect phototransformation reactions. Indeed, under the action of UV irradiation, even at 400 J/m<sup>2</sup>, the organic matter is "excited", which results in the production of triplet excited states and radical oxidants such as hydroxyl radicals, carbonate radicals and nitrogen dioxide (Canonica *et al.*, 2008). These radicals are highly oxidising chemical species that can induce transformation of organic compounds. There are also risks of reaction between 'excited' nitrite and nitrate ions and these organic compounds under UV radiation. In the presence of organic compounds, nitrate and nitrite ion photolysis, at wavelengths below 230 nm, causes the formation of unstable compounds in aqueous media (such as peroxynitrates, nitrogen oxides, hydroxyl radicals) that are likely to lead to the formation of nitrated and nitrosated organic compounds (Lee and Yoon, 2007; Vione *et al.*, 2007; Canonica *et al.*, 2008). Low-pressure mercury vapor lamps emit part of their radiation at 185 nm, when they are not properly filtered. This wavelength enables the formation of hydroxyl radicals (HO°) from water molecules. Clearly, an increase in the UV dose, or a heterogeneous dose distribution in the reactor, can increase the risk of adverse photochemical effects.

In addition, the positioning of the UV reactor in the treatment system is of vital importance: if disinfection by UV radiation were applied to water pre-treated with chlorine (Zheng *et al.*, 1999), the combination of free residual chlorine and combined chlorine in water, could promote the formation of undesirable by-products (trihalomethanes, haloacetic acids) through the generation of highly reactive halogen radicals.

Few studies have been conducted to date with medium-pressure lamps. Nevertheless, it seems highly likely that these lamps, which produce wavelengths over a wide range (200 to 600 nm), are capable of photolysing photosensitive compounds. It is therefore necessary to limit the dose delivered by this type of lamp, along with that delivered by low-pressure high-energy lamps.

#### 2.3.2.DNA repair

#### 2.3.2.1. Repair mechanisms

The DNA damage caused by UV radiation can be repaired by some microorganisms in two ways:

- Photoreactivation, which is a DNA repair mechanism, depending on the light at wavelengths from 310 to 490 nm;
- Dark-repair, a mechanism occurring independently of light.

These mechanisms are detailed in Annex C.

#### 2.3.2.2. Illustration of repair phenomena

The phenomenon of dark-repair is mostly observed when low-pressure lamps are used, irrespective of the UV dose applied, in the range from 100 to 600 J/m<sup>2</sup> (Zimmer-Thomas *et al.*, 2007). However, this phenomenon, like that of photoreactivation, is directly correlated with this dose (the reactivation rate is inversely proportional to dose applied), but also to the initial density of the microorganisms. The greater this density, the greater the reactivation levels. This is especially true when the initial concentration is greater than  $10^7$  CFU/mL.

The role of the initial density of microorganisms can be explained by (Süss et al., 2009):

- the transmittance of the medium, which is reduced when the bacterial population is increased, resulting in a decrease in the dose of irradiation received individually by each bacterium;
- DNA repair mechanisms, which may be facilitated by intercellular reactions.

A temperature effect was also noted by some authors, i.e. the phenomena of reactivation in the light or dark are favoured by a high temperature (30°C) (Salcedo *et al.*, 2007).

The photorepair phenomenon is always considerably greater than that of dark-repair and can be initiated in less than 30 minutes. The dark-repair phenomenon can be initiated in times ranging from 40 minutes (*Bacillus subtilis*), two hours (*Pseudomonas aeruginosa*), five hours (*Mycobacterium smegmatis*), and even 24 hours after the end of irradiation (*Mycobacterium tuberculosis*) (Jüngfer *et al.*, 2007).

These reactivation phenomena are heavily dependent on the microorganisms (Table VIII). Some authors suggest that these differences could be explained by varying behaviour between gramnegative and gram-positive bacteria.

Therefore, it appears that low and medium-pressure lamps are equally effective in terms of DNA damage (and thus in terms of reduction), but the damage repair phenomenon may be greater with low-pressure lamps. Thus, medium-pressure lamps might not only damage DNA, but also attack the proteins needed for repair and/or replication of the damaged DNA strands (Shin *et al.*, 2009).

Many bacteria have enzymes that can repair DNA damage in the presence of light or independently of light. Viruses do not have such enzymes, however, the genetic material of adenoviruses can be repaired by the enzymes of host cells. As for the parasite *Cryptosporidium*, Morita *et al.* (2002) highlighted the possibility of photorepair and dark-repair of DNA but with the loss of infectivity in the host animal. Similar conclusions were reached for *Giardia*, however repairs after exposure to low doses were observed.

	Compound	ε (L.mol <sup>-1</sup> cm <sup>-1</sup> ) at 254 nm, pH 7	Lamp	C <sub>0</sub> (µg/L)	400 J/m²	600 J/m²	1000 J/m²	1200 J/m²	2300 J/m²	Reference	
Pesticides	isoproturon	5944	LP (15 W)	1000	0.2%	0.3%	0.5%	0.7%	1.3%	Sanches et al., 2010	
	alachlor	543			1.0%	3.2%	5.4%	6.5%	12.5%		
	atrazine	3860			3.0%	4.5%	7.5%	9.0%	17.4%		
	diuron	16162			5.2%	7.8%	13.0%	15.7%	30.0%		
	chlorfenvinphos	8656			9.0%	13.5%	22.5%	27.0%	51.7%		
	etridiazole	720	LP (5W)	350	10.0%	-	-	-	-	Liu <i>et al.</i> , 2009	
	iopromide	-	IP	-	15%, 5.2%	-	-	-	-	Canonica	
	17-α ethinyloestradiol	-	(15W)	-	0.4%, 0.7%	-	-	-	-	et al.,	
	sulfamethoxazole	7345	MP	-	15%, 7.4%	-	-	-	-	2008	
	diclofenac	3465	(15000)	-	27%, 26%	-	-	-	-		
	ucioienac	-		5 to 137	-	-	-	90%*	97%	Kim and Tanaka, 2009 *Kim <i>et</i> <i>al.</i> , 2009 (more thop 20	
	carbamazepine	6072	LP (8W)		-	-	-	-	8%		
Dharmassutias	ketoprofen	15155			90%*	-	-	-	97%		
Pharmaceutical products	antipyrine	6626			-	-	-	-	83%		
	isopropylantipyrine	7255			-	-	-	-	78%		
	fenoprofen	800			-	-	-	-	70%		
	naproxen	3961			-	-	-	-	22%	compounds	
	indomethacin	14848			-	-	-	-	20%	monitored	
	acetaminophen	4218	218		-	-	-	-	17%	)	
	mefanimic acid	4633			-	-	-	-	9%		
	ethenzamide	743							4%		

Table VII: Examples of UV photolysis yields of some organic compounds depending on the dose applied

Microorganism	Type of lamp	UV dose (in J/m <sup>2</sup> )	N₀ (CEU/mL)	Value after irradiation (in log)	Type of repair	Value after repair (in log)	Minimum time of repair	Remarks	Reference
Escherichia coli	LP	50		4.5 (±0.2)	L	3.4 (±0.1)	30 minutes	The repair phenomenon was only monitored for 4h. The initial microorganism concentration was imprecise	Zimmer-Thomas <i>et al</i> ., 2007
					D	1.4 (±0.0)	240 minutes		
		80		5 1 (+0 2)	L	2.9 (±0.2)	30 minutes		
				0.1 (±0.0)	D	0.7 (±0.1)	240 minutes		
		200		$51(\pm 01)$	L	0.4 (±0.0)	240 minutes		
			$10^7$ to $10^9$	5.1 (± 0.1)	D	-0.2 (±0.2)	240 minutes		
O157:H7		400		5 3 (±0 1)	L	0.5 (±0.2)	240 minutes		
				0.0 (±0.1)	D	-0.3 (±0.3)	240 minutes		
	MP	50		6.4 (±0.2)	L	0.7 (±0.1)	240 minutes		
					D	0.1 (±0.1)	240 minutes		
		80		6.6 (±0.4)	L	0.9 (±0.2)	240 minutes		
					D	0.3 (±0.3)	240 minutes		
	LP	400	6.4.10 <sup>5</sup>	4.1 (± 0.1)	L	0 (±0.1)	8h	The repair was carried out in increments: it started after 8h, stalled then restarted after 36h	Süss <i>et al.</i> , 2009
			1.1.10 <sup>7</sup>	5.9 (±0.3)	L	1.2 (±0.2)	8h		
			3.4.10 <sup>7</sup>	7.1 (±0.1)	L	2.7 (±0.1)	8h		
			5.9.10 <sup>7</sup>	4.0 (±0.2)	L	3.7 (±0.1)	8h		
Pseudomonas aeruginosa		600	6.4.10 <sup>5</sup>	$5.4 \pm 0.2$	L	0 (±0.1)	8h		
			1.1.10 <sup>7</sup>	6.3 (±0.5)	L	1.6 (±0.3)	8h		
			3.4.10 <sup>7</sup>	7.5 (±0.2)	L	3.0 (±0.2)	8h		
			5.9.10 <sup>7</sup>	5.0 (±0.1)	L	2.4 (±0.1)	8h		
		100-600			0		>6h		Jüngfer <i>et al.</i> , 2007

Table VIII: Comparative efficacy of low-pressure and medium-pressure lamps on i) inactivation and ii) aftergrowth capacity of some microorganisms (L: in the light = photorepair; D = in the dark = dark-repair)

Enterococcus faecium	LP	400	3.3.10 <sup>6</sup>	6.3 (±0.1)	L	0 (±0.1)	>66h		Süss <i>et al.</i> , 2009
			1.2.10 <sup>7</sup>	5.9 (±0.2)	L	0 (±0.1)	>66h		
			4.4.10 <sup>7</sup>	4.5 (±0.1)	L	0.3 (±0.3)	>66h		
			6.7.10 <sup>7</sup>	3.5 (±0.2)	L	0.6 (±0.2)	>66h	No significant	
		600	3.3.10 <sup>6</sup>	7.1 (±0.2)	L	0.3 (±0.2)	>66h	photoreactivation	
			1.2.10 <sup>7</sup>	7.3 (±0.2)	L	0.9 (±0.2)	>66h		
			4.4.10 <sup>7</sup>	5.0 (±0.3)	L	-0.7 (±0.2)	>66h		
			6.7.10 <sup>7</sup>	3.7 (±0.1)	L	-0.2 (±0.4)	>66h		
		100-600			D		>6h		
Caulobacter crescentus	LP	100-600			D		<2h		Jüngfer <i>et al.</i> ,
Aquabacterium commune	LP	100-600			D		<2h	No dark-repair if UV dose >200 J/m <sup>2</sup>	2007
Bacillus subtilis	LP				D		>40 minutes		
Mycobacterium smegmatis	LP				D		> 5h		Papavinasasund aram <i>et al.,</i> 2001
Mycobacterium tuberculosis	LP				D		> 24h		
Mycobacterium terrae	LP/MP	50-400			L		<30 minutes	No dark-repair irrespective of the UV dose applied	Bohrerova et Linden, 2006
Coliphage MS2	LP/MP				L	0		No reactivation	
					D	0		possible for MS2	Shin <i>et al.</i> , 2009
Adenovirus2	LP	300-900			L	1.0		Reactivation observed	
	MP				L	0.0		only with LP lamp	

## 2.4.Conclusions

Disinfection treatment by UV radiation requires, depending on the classes of microorganisms, radiation doses in the range of 300 to 400  $J/m^2$ . If the dose is lower, microorganisms will not be "killed" and may be able to repair themselves. At 400  $J/m^2$  an inactivation efficacy of approximately 4-log is recognised with respect to bacteria and protozoa. However, for viruses, there may be greater resistance as, for example, in the case of adenoviruses which require radiation doses in the range of 1500 to 1600  $J/m^2$  to achieve a 4-log reduction.

In addition, this radiation paired with other oxidants can induce secondary reactions: UV-ozone, UV-hydrogen peroxide, UV-chlorine dioxide. The UV-hyperchlorite pairing generates singlet oxygen and highly reactive halogenated free radicals that also result in secondary reactions.

Therefore, these paired treatments are not approved for producing water intended for human consumption, with the exception of groundwater containing only chlorine solvents with one or two carbon atoms.
# 3. Normative and regulatory context

French, European and third country regulations as well as the associated standards are very important with regard to UV radiation disinfectant treatment of water intended for human consumption. Annex D lists the various standards and regulations that have been adopted for the assessment and marketing of UV reactors.

# 3.1.Conditions for placing UV reactors for the treatment of WIHC on the French market

#### 3.1.1.Regulations applicable to UV reactors

Low-pressure-type UV reactors are included in the Circular of 28 March 2000 among the groups of treatment products and systems that can be placed on the market for disinfecting WIHC. This circular refers back to Circular DGS/PGE/1-D no. 52 of 19 January 1987 regarding recommended conditions of use. These systems are placed on the market without individual review or approval and UV radiation treatment equipments can be marketed for the treatment of WIHC when the UV lamps used are of the low-pressure type and the radiation dose is at least 250 J/m<sup>2</sup>. It should be noted that under these conditions, use applies to traditional bactericidal disinfection treatment and these conditions of use do not emphasise any *de facto* efficacy with respect to parasites (*Cryptosporidium* and *Giardia* in particular), or viruses.

However, the provisions established at that time are now partly outdated and if a manufacturer wishes to market a UV reactor that is:

- fitted with medium-pressure mercury vapor lamps;
- and/or designed to inactivate parasites or viruses;

then this reactor is considered to be an 'innovative' device in terms of the specific current provisions. Its marketing is then covered by the provisions of Article R.1321-50-IV of the French Public Health Code cited above.

# 3.1.2.History of the assessment of marketing authorisation application dossiers for UV reactors

Since its inception, AFSSA, whose missions were entrusted to ANSES as of 1 July 2010, has been responsible for evaluating marketing authorisation application dossiers for UV reactors using lamps other than low-pressure mercury vapor lamps and claiming action broader than bacterial disinfection. As of 19 April 2010, 27 requests for an opinion for approval of UV reactors were submitted to AFSSA.

# 3.2. Other normative or regulatory references

In order to determine the regulatory requirements for the marketing of UV reactors for the disinfection of WIHC in European Union Member States, a questionnaire was sent to the representatives of Member States participating in the ENDWARE group. Ten countries responded, including Austria, the United Kingdom and Ireland, which have established specific requirements for the marketing of UV reactors.

Other countries also have systems for qualifying UV reactors for marketing, they include Norway, Switzerland, New Zealand, the USA and Canada.

Finally, the WHO, in its guidelines on the quality of water intended for human consumption, proposes doses of UV radiation for inactivating various microorganisms.

All of these are listed in Annex D.

# **3.3.Normative context**

Individual country provisions are based on the following documents:

- Germany: DVGW Technical Standard W294 (1 to 3) entitled: UV Systems for Disinfection in Drinking Water Supplies; part 1: qualitative, functional and operation requirements; Part 2: qualitative, functional and disinfection-efficacy testing; Part 3: analysis windows and sensors for radiometric monitoring of UV disinfection equipment; requirements, testing and calibration (June 2006);
- Austrian standard: ÖNORM M 5873-1 and -2 (MP and LP) entitled: Plants for the disinfection of water using Ultraviolet radiation – Requirements and testing; Part 1: low-pressure mercury vapor lamp plants (March 2001); Part 2: medium-pressure mercury vapor lamp plants (August 2003);
- USA guide: National Water Research Institute (NWRI) and American Water Works Association Research Foundation (AWWARF) entitled "Ultraviolet Disinfection Guidelines for Drinking Water and Water Reuse" (Second Edition 2003);
- American Standard NSF-ANSI 55 entitled: Ultraviolet Microbiological Water Treatment Systems (2007);
- US EPA protocol entitled Ultraviolet Disinfection Guidance Manual (UVGM) for the Final Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR) (Updated in November 2006).

The European Standard EN 14897/IN1 +A1 for water conditioning equipment inside buildings has not been taken into consideration, although it is very similar to the requirements of European, German and Austrian standards.

# 4. Evidence of safety and efficacy

# 4.1.Safety of systems using UV reactors

Materials coming into contact with water during its passage through the UV reactor must not allow undesirable molecules to migrate and degrade the water's quality. Requirements are necessary for all treatment materials or media coming into contact with water. Furthermore, in the case of UV reactors, materials are subject to conditions that are not typically encountered during water distribution. Thus, evidence of compliance for organic materials does not include migration tests while being subjected to UV radiation and high temperature.

Currently, only stainless steel is used to make the UV reactors' casing. If another material is used, it must be proven to be inert to UV radiation.

Products used for maintaining the reactors should not cause degradation of water quality. The products and cleaning systems currently used for reactor maintenance are:

- Continuous or periodic chemical cleaning with the following products:
  - phosphoric acid;
  - white vinegar (just for removing calcium carbonate);
  - o dicarbolic acid in acid medium.
- Automatic mechanical cleaning.

A flushing procedure should be defined and evidence of the efficacy of the flushing should be provided.

To ensure that the wavelengths responsible for generating hazardous by-products are properly filtered out by the doped quartz sleeve, the emission spectra of the lamp alone and fitted with its sleeve should be provided. Moreover, if the doped quartz or the filter come in contact with the water, it must be ensured that the doping element is known and that it is not capable of migrating into the water during contact.

## 4.1.1.Photonic modelling

Photonic modelling (see Annex A §2) can be used to determine the profile of the distribution of the dose that the particles receive in the reactor. This distribution helps estimate the percentage of by-products formed that are deemed to be undesirable.

When using models available on the market, the modeller must enter certain data, including the number of particles (comparable to microorganisms) and transmittance (that is, of water and quartz), which are essential parameters. The choice of turbulence model (Liu *et al.*, 2007) also has an impact on the predictions of inactivation and reduction equivalent dose (Munoz *et al.*, 2007). The current models do not take into account the aging of the lamps and the formation of deposits on the sleeves, which depend on the physico-chemical composition of the water.

In any case, the computational fluid dynamics (CFD) methodology should not be used to verify the disinfection efficacy of a UV reactor. This modelling step should be restricted only to the design stage of the reactor, even if some authors indicate that modelling would provide information on both the design of the reactor and on the efficacy of the system.

Only biodosimetric tests can validate the efficacy of a UV reactor.

This is emphasised by Pan and Orava, 2007, who indicate that "CFD simulations may not be a sufficiently accurate tool for predicting the flow rates in a UV reactor because the *electrical efficacy* 

(defined as the electrical energy actually usable for inactivation of bacteria) is uncertain with the current models, and that only the *mechanical efficacy* of the reactors can be predicted from CFD simulations".

# 4.2.Evidence of efficacy

### 4.2.1.Standards requirements

The recommendations in terms of efficacy are the following for the three existing standards:

- US EPA (USA), which uses biodosimetry tests with MS2 bacteriophage as a challenge microorganism. In its Surface Water Treatment Rule (SWTR) it establishes the objective of 400 J/m<sup>2</sup> for a 4-log reduction in *Cryptosporidium* and *Giardia* and 0.5-log in viruses (adenovirus);
- DVGW 2006 (Germany), which requires that the disinfection efficacy of a UV reactor corresponds to a Reduction Equivalent Dose (RED) of at least 400 J/m<sup>2</sup> related to a wavelength of 253.7 nm;
- The ÖNORM M 5873-1 and -2 standards (Austria), which require, like the German standard, that a UV reactor deliver a RED of 400 J/m<sup>2</sup> at a wavelength of 253.7 nm. At this dose, a reduction of at least 4-log in pathogenic bacteria, viruses and protozoa transported by water is assured in accordance with the state of the science.

#### 4.2.2.Test protocols

The radiation dose required for inactivation of microorganisms cannot be determined *a priori* because it depends on the quality of the water to be treated (particularly turbidity) and the geometric and hydrodynamic configurations of the reactor used. That is why it is determined experimentally by biodosimetry, which is a standardised method using challenge microorganisms whose sensitivity to UV radiation has been calibrated.

According to the standards, the UV equipment used for the disinfection of WIHC must offer disinfection potential corresponding to a UV dose of at least 400  $J/m^2$ . This should be verified with a microorganism presenting a 4-log reduction for a dose of 400  $J/m^2$  in static irradiation. However, microorganisms that are easy to produce and safe when handled in the laboratory are not always suitable for verifying these reductions.

#### Biodosimetry principle:

Experimental determination of the RED is performed in two steps:

- establishment of a response curve at the dose of UV radiation for a test microorganism or challenge microorganism, chosen based on its resistance, in controlled static exposure conditions;
- establishment of the RED after exposure of this microorganism in the UV reactor under dynamic conditions. The RED is the dose for which the same level of inactivation is obtained in full-scale testing as under static laboratory conditions.

The validated dose is obtained by applying a corrective factor to the RED which takes into account experimental biases and uncertainties.

#### 4.2.2.1. Choice of challenge microorganisms

Disinfection using UV radiation is recommended in particular for faecal protozoa with resistant dissemination forms such as *Giardia* cysts, *Cryptosporidium* oocysts or viruses. Handling these pathogens, in high concentrations, may be hazardous for the operators during validation tests, hence the use of non-pathogenic microorganisms likely to be found in the water, and whose sensitivity to UV radiation is similar to the target microorganisms. These test microorganisms should be easy to produce in large quantities, culture and count quickly and have stable characteristics over time. It is also necessary to consider the type of inactivation curve of the microorganism which may be diphasic with an initial latency phase (photochemical repair after low dose irradiation or the concept of inactivation by multiple sites or multiple strikes). Sensitivity to UV radiation must be assessed in the linear part of the inactivation curve ranging between the changes in slope, which is the case for *Bacillus subtilis* spores, for example.

Among the candidate microorganisms, MS2 bacteriophages and *Bacillus subtilis* spores show significantly greater resistance than those of *Giardia* and *Cryptosporidium* (Table IX).

Table IX: List of microo	organisms recommen	nded for assess	sing the disinfe	ction efficacy	of UV	radiation
reactors and their sensi	itivity to UV radiation	(from US EPA,	2006)			

Microorganism	Dose UV (J/m <sup>2</sup> ) to reach the indicated log reduction				
Microorganism	1 log	2 log	3 log	4 log	
Bacillus subtilis	280	390	500	620	
MS2 phage	160	340	520	710	
Qß phage	109	225	346	476	
PRD-1 phage	99	170	240	300	
B40-8 phage	120	180	230	280	
φx174 phage	22	53	73	110	
E. coli	30	48	67	84	
T7 phage	36	75	118	166	
T1 phage	≈ 50	≈ 100	≈ 150	≈ 200	

The standards recommend using the following test microorganisms:

- DVGW: spores of Bacillus subtilis ATCC 6633;

- ÖNORM M 5873 1 and 2: spores of Bacillus subtilis ATCC 6633;
- US EPA 2006: bacteriophage MS2 ATCC 15597-B1.

#### 4.2.2.2.Static laboratory tests: standardisation using the challenge microorganism

The challenge microorganism's sensitivity to UV radiation is determined in static mode, under defined laboratory conditions, using a collimated beam apparatus that generates precisely measured radiation.

This device has a horizontal casing made of non-reflective opaque material with a calibrated UV radiation source and a known emission spectrum. Typically, this is a low-pressure UV radiation lamp emitting at 254 nm. The source must emit constant radiation, which requires a voltage stabiliser or an electronic control device. The radiation emitted by the source passes through a collimator tube, made of non-reflective opaque material, facing down. It is directed onto a suspension of microorganisms contained in a cylindrical cup-like Petri dish in UV ray-absorbing material to avoid reflections. The light field of the collimated beam covers the entire surface of the cup and is perpendicular to the level and unagitated surface of the sample. The height of the liquid is between 0.5 and 2 cm.

Standardising the sensitivity of the test microorganism is carried out with the same water as that used in the full-scale tests. The two types of tests, static and dynamic, are conducted on the same day to minimise variations related to the production of the microorganism.

The UV energy is measured by a reference radiometer.



Figure 15: Laboratory irradiation equipment diagram (ÖNORM, 2001)

The inactivation curve of the test microorganism, based on the UV radiation received, is obtained by exposing the suspensions, in concentrations of about  $10^6$  to  $5.10^6$  PFU (for viruses) or CFU/mL (for bacteria) and distributed into the Petri dishes. The suspension transmittance must be greater than 90% at 254 nm measured in a 1 cm long quartz cell. For the assessment, only dishes with colony numbers between 20 and 200 should be counted.

At constant energy (W/m<sup>2</sup>), exposure time (s) is modified to cover an expanded exposure area from 100 to 800  $J/m^2$  with at least six to eight doses.

$$D(J/m^2) = I(W/m^2) \times t(s)$$

From the experimental data

- doses of exposure to UV radiation in J/m<sup>2</sup>,

and

- the reduction factor: log  $(N/N_0)$  where  $N_0$  and N are the concentrations of microorganisms before and after exposure to UV radiation,

the inactivation curve of the microorganism is derived from the curve of the reduction factor depending on the dose. The linear part of the inactivation curve is described by the following formula:

$$\log (N/N_o) = d + kH$$

whered: point of intersection of the line with the y axis

k: slope of the line (m<sup>2</sup>/J)

H: UV dose (J/m<sup>2</sup>)

This inactivation curve at a wavelength of 253.7 nm is validated if it is within the tolerance limits defined by the abovementioned standards.



Figure 16: Admissible region for biodosimetric inactivation curve (ÖNORM, 2001)

In addition, data from inactivation of the challenge microorganism must satisfy the following criteria:

The inactivation curve obtained serves as a benchmark for the dynamic tests described below.

#### 4.2.2.3.Dynamic tests on the pilot bench

This is a full-scale check to ensure that a defined dose (for example 400 J/m<sup>2</sup>) is actually delivered by the UV reactor and to establish the reactor's conditions of use. The parameters taken into account are the flow rate and UV transmittance of the water, the power and condition of the lamps, UV energy, fouling, the hydraulic configuration of the pipelines and of the reactor.

The UV reactor is placed in a pilot bench on an industrial-scale site or in a specialised facility. Water stored in a reservoir upstream is distributed in a system as shown in Figure 17.





Drinking water quality is used that has high UV transmittance (99%), water turbidity of less than 0.1 FNU, and iron and manganese concentrations both less than 10  $\mu$ g/L. If necessary, the residual chlorine is neutralised by sodium bisulphite in order not to affect transmittance.

The UV absorbers used to modify the UV transmittance of the water are:

- freeze-dried coffee,
- lignin sulfonate,
- humic acids.

The lamps fitted to the reactor must have been operating for at least one hundred hours before testing.

The protocol includes performing two types of tests "H" and "L", each conducted under conditions of maximum, intermediate, and minimum flow:

- the "H" test is performed at the lamp's maximum power by decreasing UV transmittance by adding an absorber until a given energy is obtained, which is the minimum irradiance for the lamp's maximum power,
- the "L" test is performed at high UV transmittance by varying the power of the lamp, which is the minimum irradiance for the highest UV transmittance by reducing the power of the lamp. The emission of UV radiation is set to a maximum of 70% of the radiant power of a new emitter.
- for a pair of characteristic parameters minimum irradiance and maximum flow it is necessary, with two variations for setting the minimum irradiance (test "H" and "L"), to obtain a reduction equivalent dose (RED) of at least 400 J/m<sup>2</sup> for the biodosimetric analyses.

Each type of test is carried out in duplicate, in two independent sequences.

The test microorganism or challenge microorganism is introduced upstream of the reactor, under defined operating conditions, in order to obtain a concentration of  $10^6$  to  $10^7$  CFU/L. Water samples are taken before(N<sub>0</sub>) and after (N) the UV reactor at a rate of three to five replicas per test condition at one minute intervals. Each replica is analysed in triplicate. The arithmetic mean of the counts related to the dilution factor is converted into a decimal logarithm. The result is obtained by averaging the logarithmic mean of the replicas for which the standard deviation must not exceed  $\pm$  0.2. The calculation of N/N<sub>0</sub> determines the reduction in the challenge microorganism obtained for each test condition.

#### The reduction equivalent dose (RED)

The RED is obtained by transferring to the dose-response calibration curve obtained in static situations, the reduction in test microorganism obtained in dynamic situations.

The RED of each replica for each operating condition can be calculated using the equation of the dose-response relationship previously developed.

$$RED = -\frac{1}{k} \times \log \left[ 1 - \left( 1 - \frac{N}{N_0} \right)^{10^{-d}} \right]$$

 $\frac{N}{N_0}$  = biodosimetric inactivation rate

k = loss of sensitivity to UV (m<sup>2</sup>/J)

d = distance between the ordinate at the beginning of the curve and the zero in the inactivation curves of the microorganisms

Test point number	1	1*	2	2*	3	3*	
Parameter to be varied	UV transmission factor	Lamp power	UV transmission factor	Lamp power	UV transmission factor	Lamp power	
Test settings	Irradiance $E_1$ at flow rate $q_1$		Irradiance $E_2$ at flow rate $q_2$		Irradiance $E_3$ at flow rate $q_3$		
RED for the first run, J/m <sup>2</sup>	405	420	401	425	408	402	
RED for the second run, J/m <sup>2</sup>	417	403	413	416	410	424	



Figure 18: Example of representation of the admissible operating range: minimum irradiance and maximum flow (ÖNORM, 2001)

The conditions of use of the UV reactor to deliver a RED of 400 J/m<sup>2</sup> are obtained from the following two types of relationships:

- admissible flow rate depending on the UV transmittance,
- minimum lamp power to comply with, depending on the flow rate.

# 5. Emerging technologies and technological outlook

UV irradiation is undergoing constant development. Low-pressure and medium-pressure mercury vapor lamps are the most frequently used at the current time, but new technologies are being explored. For example, light emitting diodes (LEDs) that emit at 365 nm (Mori *et al.*, 2007), hollow cathode lamps that emit at 220 nm or less (Soloshenko *et al.*, 2006), excimers discharging in a mixture of inert gas (xenon, krypton) and halogen (bromine, chlorine) which emit at a wavelength of about 280 nm (Xe Br) (Naunovic *et al.*, 2008), at 308 nm (Xe CI) and even at 222 nm (Kr CI) (Sosnin *et al.*, 2006) are all being developed.

Studies of these devices relate only to their performance in terms of bacterial inactivation efficacy without addressing the risk of formation of by-products that could be toxic. A summary of the literature available on the subject is presented in Annex E.

# Conclusion

In France, as well as in other European countries and more broadly at the global level, validation of systems for the disinfection of water intended for human consumption using UV reactors is a prerequisite to their being placed on the market.

Growing interest in treating water by UV radiation, in addition to its bactericidal action, is based primarily on its potential for inactivating protozoa of the genera *Cryptosporidium* and *Giardia*.

However, disinfection by UV radiation has some limitations. Its lack of a residual effect does not ensure the preservation of the microbiological quality of the water being distributed. Microorganism repair phenomena can occur and cause aftergrowth. The virucidal efficacy varies greatly depending on the nature of the virus (DNA or RNA). To control the critical points of this disinfection system a minimum dose of irradiation must be received by the water and, especially in water that might contain viruses, chemical disinfection treatment must be coupled with UV radiation treatment.

It should be remembered that the UV reactors placed on the market have been qualified for their safety and efficacy and these properties are guaranteed provided that the reactor:

- is operated under the defined conditions of use (flow/transmittance of water) after efficacy tests have been performed and ensuring that the treated water is actually receiving a given dose of irradiation;
- is in all respects identical to the model on which the efficacy tests were conducted (in terms of reference of the lamp, the cut-off sleeve, materials, etc.);
- is properly used, maintained and inspected by the individual in charge of water production.

The guidelines outlined below are aimed at improving the assessment of safety and efficacy of treatment of water intended for human consumption by UV reactors. They give to those responsible for marketing these systems a detailed technical document to constitute their authorisation dossier in France, by clarifying and explaining the points on which the assessment is based. The approach integrates the international context and takes into account the standards applied in other European countries to facilitate the process of mutual recognition. Their scope of application only covers reactors implementing low- and medium-pressure mercury vapor lamps positioned in the flow of water to be treated. Thus, these guidelines should be adapted where necessary for the assessment of UV systems using other types of lamps, such as diodes or flash systems that emit a very short signal but with very high photon energy, or systems in which the lamps are placed outside the water flow and which transfer the UV radiation. They should also be modified if the progress of scientific knowledge warrants it.

ANSES recommends that a list of authorised reactors be kept up to date and made public.

In addition, ANSES emphasises the importance of conducting a more precise study of this type of system for disinfection uses with other types of water, particularly water used in the food industry, and seawater used in fish auctions or shellfish culture, to ensure that the technical complexity and mastery of the system are properly considered.

Finally, the evidence presented in the report confirms the need for further knowledge about the distribution of the UV radiation dose in the flow of treated water, the conditions of by-product formation and microorganism repair phenomena. The latter point also concerns the field of basic and targeted research.

ANSES takes responsibility for the report, its conclusions and guidelines issued by the collective expert appraisal conducted within the dedicated Working Group and validated by the Expert Committee (CES) on Water.

The Director General

Marc MORTUREUX

# Guidelines for the assessment of reactors fitted with ultraviolet lamps and the efficacy of these systems for the disinfection of water intended for human consumption

# **1. Safety and efficacy**

## 1.1. Safety of materials

The applicant shall state the different parts of the reactor, the materials from which it is made and specifies the percentages of contact with water.

All materials must have proof of compliance with health practices in force (health compliance certificate (ACS) or compliance with French positive reference lists (CLPs)).

For organic materials, if the contact area is greater than 5% of the total wetted surface, the applicant shall provide proof that it does not react to UV radiation, as the ACS does not include verification of this parameter.

The life cycle of each component shall be specified, especially the method for recycling and processing mercury vapor lamps.

## 1.2. Safety of maintenance products

The applicant shall list the maintenance products and protocols that it recommends for the reactors and the cut-off sleeves. These protocols must specify:

- If the maintenance product is used outside of the production phases (during shutdowns): methods of flushing and monitoring the efficacy of the flushing must also be described.
- If the maintenance product is used during production (uninterrupted): the applicant must then demonstrate that the product has no impact on water quality.

Products must show proof of compliance with a valid health certificate (CLP).

It is recommended that an inventory be made of the products used.

## **1.3.** Risk of formation of undesirable by-products related to lamp emissions

The applicant shall provide the spectrum of the lamp alone or, as applicable, the spectrum of the lamp fitted with a cut-off device.

It shall also provide the percentage of irradiance emitted in the UV-C spectrum.

On this point, two requirements are listed in the following standards:

- ÖNORM M 5973 Standards -1 and -2 in their Article 6.3, which requires:
  - that irradiance at wavelengths below 240 nm be less than 3% of the irradiance measured between 240 and 400 nm,
  - AND, for medium-pressure lamps, that irradiance emitted at wavelengths other than 253.7 nm not equal more than 85% of the irradiance emitted over the entire field of UV-C radiation.
- OR, DVGW Standard in its Article 9.3, which requires that irradiance at wavelengths below 240 nm correspond to less than 5% of the total irradiance emitted between wavelengths from 240 to 290 nm.

The Working Group suggests following energy standards requirements:

- For low-pressure lamps: that irradiance at wavelengths below 240 nm corresponds to less than 5% of the total irradiance emitted between wavelengths from 240 to 290 nm;
- For medium-pressure lamps: that irradiance at wavelengths below 240 nm corresponds to less than 3% of the total irradiance emitted between wavelengths from 240 and 400 nm.

## 1.4. Risk of formation of undesirable by-products related to dose

The applicant shall provide the dose distribution within the reactor and the dose received by the flow of water, both obtained at different reactor operation flow rates.

The following criteria of acceptability are proposed:

- For low-pressure lamps: the RED shall be 600 J/m<sup>2</sup> with at least 90% of the water flow receiving a dose of between 400 J/m<sup>2</sup> and 1000 J/m<sup>2</sup> and less than 5% of the flow receiving a dose of less than 400 J/m<sup>2</sup> (to limit dark-repair phenomena) and less than 5% of the flow receiving a dose greater than 1000 J/m<sup>2</sup> because there is only one emission wavelength and thus the risks of by-products forming with high energy are limited.
- For medium-pressure lamps: the RED should be 400 J/m<sup>2</sup> with at least 90% of the water flow receiving a dose of between 400 J/m<sup>2</sup> and 800 J/m<sup>2</sup> and less than 5% of the flow receiving a dose of less than 400 J/m<sup>2</sup> (to limit dark-repair phenomena) and less than 5% of the flow receiving a dose greater than 800 J/m<sup>2</sup> because there are several wavelengths emitted and as a result the high limit is lowered to minimise the risks of by-products forming with high energy.

## 1.5. Risk of formation of undesirable by-products related to precursors

The applicant shall present test results on water loaded with 50 mg/L of nitrates and provide evidence that under different operating conditions the concentration of nitrites after treatment is less than 0.1 mg/L in the water produced. This test is not required for reactors fitted with low-pressure/low-energy lamps.

#### **1.6.** Quality of the water to be treated:

- To prevent the formation of undesirable by-products:
  - the water to be treated should not contain oxidants (ozone, chlorine, chlorine dioxide, chloramine and potassium permanganate);
  - when the water contains traces of iodine at a concentration above 10 μg/L, the risk of forming sapid iodised compounds cannot be excluded.
- To prevent the formation of deposits on the sleeves that reduce the transmission of UV radiation and on the radiometers:
  - the water must have a calcium carbonate equilibrium at 40°C (thus slightly aggressive at ambient temperature) to avoid the potential precipitation of calcium carbonate;
  - o the iron concentration in the water to be treated must not exceed 50 μg/L;
  - $\circ$  the manganese concentration in the water to be treated must not exceed 20 µg/L.

In all cases, the cleaning methods must be adapted to the quality of the water to be treated.

- To ensure effective transmission of the UV radiation in the water to be treated:
  - $\circ$  the turbidity of the water entering the reactor must not exceed 0.3 FNU;
  - its UV absorption must not exceed 10 m<sup>-1</sup> at the wavelength of 253.7 nm.

## 1.7. Efficacy of disinfection

Efficacy is monitored indirectly by biodosimetry. The applicant shall provide biodosimetric test results which consist of two steps:

- a static test that provides a correlation between the UV radiation dose and inactivation rate of a challenge microorganism, the latter being either an MS2 bacteriophage (as recommended by the US EPA), or spores of *Bacillus subtilis* (in accordance with European standards).
- a semi-dynamic test by varying the operating parameters (flow rate, transmittance of the water and power of the lamps) using the same challenge microorganism as for the static test to obtain graphs for defining the reactor's scope of application to ensure a RED of 400 J/m<sup>2</sup> or 600 J/m<sup>2</sup>.

These tests can be performed according to existing standards: ÖNORM M 5873-1 or -2, DVGW W294 or US EPA UVGM. They are carried out by certified test centers according to these standards or equivalent Good Laboratory Practices (GLP) (Section 4.5.2 of the DVGW W294-2 Standard). If testing is conducted outside the European Union, proof of the laboratory's competence and independence shall be provided.

In all cases, the applicant shall submit the entire protocol and all test results in the authorisation dossier.

Each reactor model shall be identified by its name, configuration, the number of lamps, and the existing references of the lamp and the sleeve, if any. The tests are conducted for each model in the same range and with the type of lamp and sleeve sold.

## **1.8.** Monitoring of operating conditions

The applicant shall provide the reference, the characteristics and origin of the reference radiometer that it is using. It shall also specify: measurement uncertainty, procedures for verifying calibration, calibration procedures (frequency, competence of the operator, etc.).

The applicant shall provide the origin and reference for the working radiometers it uses. It shall also specify the admissible tolerance with the reference radiometer, the frequency of verification with the reference radiometer, and the measurement uncertainty.

The applicant shall specify and justify the number of radiometers in operation compared to the number of reactor lamps. It shall indicate and justify the position of the radiometers with respect to the reactor configuration.

In all cases the working radiometers should measure the dose in the water flow.

## 1.9. Certification period

Certification should be issued for a maximum period of five years. This time period is identical to that applied for German and Austrian certificates.

# 2. Recommendations for use and monitoring

#### 2.1. Home use

Domestic use of reactors fitted with UV radiation lamps (UV reactors) is not recommended, mainly because of the time required to heat the lamps and the risk of formation of secondary products when treating chlorinated water. According to current knowledge, the biocidal effect only appears after the lamps have been heated for a period of five to ten minutes. As a result, intermittent or sequential use of these treatments is strongly discouraged. Furthermore, the continued use of these systems on non-circulating water or water in a closed loop results in warming of the water and the risk of microbiological growth in the system. Past experience has shown that the use of UV reactors in domestic plumbing systems is strongly discouraged for water from:

- public supplies going to private parts of the system,
- private wells for single-family use,
- private wells with provisions for public water use,
- drinking water coolers,
- hot water.

Only treatment for the following types of water can be recommended as part of industrial systems in plants for water:

- used for the production of water intended for human consumption,
- used in the food industry in the manufacture of food or in contact with food. This water comes from the water supplied by the public water system or produced from a private resource in accordance with the French Public Health Code (CSP).

#### 2.2. Role of UV radiation treatment in the drinking water treatment chain

Given the quality of water necessary to ensure the optimum safety and efficacy of UV radiation treatment, it is recommended that the UV reactor be placed at the end of the chain.

Because of the inactivation rate of some viruses, it is recommended that a disinfection step be added using an approved biocidal product before the UV radiation treatment. Depending on the viral load of the water, the water production manager must ensure that the chain has the capacity for the disinfection required.

The system of disinfection using UV radiation has no residual effect and if such an effect is sought, particularly for the transport of water, an approved remanent biocidal product should be implemented.

#### 2.3. Monitoring precautions

In addition to UV radiometers, the UV device is fitted with sensors to ensure the quality of the water before and after UV radiation treatment (temperature, turbidity, etc.) and sensors to monitor the operating parameters of the reactor (flow rate, electricity consumption, etc.).

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# Annex A: Photonic and hydraulic modelling

# 1. Computational Fluid Dynamics model

The ongoing inactivation of microorganisms in UV reactors is particularly dependent on hydrodynamism and mixing conditions within the irradiated part of the reactor. Models based on thoroughly agitated open reactors or open piston flow reactors with axial dispersion were developed initially (Severin *et al.*, 1984, Scheible, 1987). Tracers were also used to determine the dwell times within these reactors (Qualls and Johnson, 1985).

The current trend is to use Computational Fluid Dynamics (CFD) models. In these CFD analyses, the first step is to determine the velocity vector field by solving the Navier-Stokes motion equations (Munoz *et al.*, 2007) in conjunction with an appropriate turbulence model like the  $k - \varepsilon$  model, or the Reynolds stress model or a combination of the two: Reynolds-averaged Navier-Stokes equation (or RANS equation) model (Sozzi and Taghipour, 2006):

# $\nabla (\rho, \vec{\upsilon}) = -\nabla P + \nabla (\vec{\tau}) + \rho g$

where:

- ρ, density;
- velocity;
- P, pressure;
- $\tau$ , Reynolds stress; and
- g, gravity.

In general, manufacturers use commercial software to solve this equation (FLUENT, for example).

Once the velocity field has been determined, the whole reactor is discretised: representation of the reactor in the form of more or less structured calculation meshes, and in a varying number, depending on the size and shape of the reactor (mesh-generation software: GAMBIT, for example). The volumetric reaction rates are calculated based on the local fluence rate and on the concentration of microorganisms in each mesh. Each mesh is treated as a thoroughly agitated homogeneous reactor.

Based on this step, two distinct approaches can be used to determine fluence rates: the Eulerian approach (Elyasi and Taghipour, 2006) and the Lagrangian approach (Sozzi and Taghipour, 2006).

Irrespective of the method used, the modeller must consider all the input variables for the model, and primarily, the number of particles used to simulate the microorganisms. These data take on a prominent character in the three-dimensional computational fluid dynamics (3D-CFD) models used to simulate high-volume, multi-lamp UV reactors where a large number of finite volume elements is required to describe the reactor accurately.

#### 1.1 Lagrangian approach

In this approach, also called the particle tracking approach, microorganisms are treated as discrete particles, and the likely routes of these particles within the reactor are calculated either by solving an equation of the amount of particle motion (p = mv, where p is the amount of motion, m the particle mass, v the velocity), or by using a random-walk algorithm. Accumulated doses of UV radiation received by each microorganism particle are then determined by numerical integration of the fluence rate field and particle trajectory information. By repeating this calculation for numerous particles, a UV radiation dose distribution is produced. The digitised dose distribution can be combined with kinetic

models of microorganism inactivation by UV radiation to generate a Reduction equivalent dose (RED) for a given microorganism.

Particle trajectories are simulated on the basis of a continuous and homogeneous velocity field within the reactor. A statistically representative number of particles (representing the microorganisms) is introduced at the reactor inlet (N<sub>0</sub>) and the dose of UV radiation absorbed is integrated along the trajectory of each of the particles. The dose absorbed at each point is calculated by multiplying the mean fluence rate (E) at a given point of the reactor for the time  $\Delta t$  that the particle is subjected to this localised fluence rate. For each dose interval i, the number of live microorganisms N<sub>i</sub> is calculated as follows:

where:

$$N_i = \alpha_i \times N_0 \exp(-kD_i)$$

- $\alpha_i$  = the fraction of particles that receive the dose D<sub>i</sub>;
- k, the inactivation rate constant for the reference microorganism;
- D<sub>i</sub>, the average dose for a given interval i.

The sum of all living particles over the entire range of doses gives the estimated total number of living particles (microorganisms) leaving the reactor, N:

$$N = \Sigma N_i$$

#### 1.2 Eulerian approach

In this approach, microorganisms are seen instead as reactive tracers in a chemical reactor, and inactivation is determined by the use of equations showing the convection-diffusion phenomena within the reactor, including a reaction term.

The conservation-of-species (microorganisms) equation is solved simultaneously with the transport equations. The local mass fraction of each species is predicted by solving convection-diffusion equations to calculate the concentration of living microorganisms throughout the range studied:

$$\nabla (\vec{v} C) = - \nabla \vec{J} + R$$

where  $\vec{J}$  is the diffusion flux (including the dispersion by turbulence) of the species and R is the reaction rate. For microorganisms that have a linear rate of inactivation, characterised by a rate constant k, the inactivation rate R is equal to:

$$R = -k E C$$

where E is the local fluence rate and C is the concentration of microorganisms.

#### 2. Fluence rate distribution model

#### 2.1Some concepts of optics

In a UV reactor, radiation emitted by the lamp must pass through a layer of air surrounding the lamp, then cross the quartz sleeve (doped or not doped) before reaching the water to be disinfected. The radiation emitted by the lamp is attenuated by the reflection and absorption phenomena while passing through these different media (air, quartz, water). Figure A1 describes these different factors for a typical optical path.



Another component of the refraction is the focus effect. If no refraction phenomenon is considered, the radiation power emitted from a point source within a finite difference angle equal to  $2 \Delta \theta_1$  (Figure A2) and traversing a distance equal to  $d_1+d_2+d_3$ , would cover a circle with diameter  $g_{WO}$ . If the fact that the UV lamps have a cylindrical symmetry is exploited, this cross-section becomes a truncated cone of area  $A_{WO}$ , with an angle of aperture of  $2 \theta_1$ , the lamp axis as the axis of the cone and the generatrix of this angle as  $g_{WO}$ .



By including the refraction phenomena at the air/quartz/water interfaces, while keeping the optical path length  $d_1+d_2+d_3$  constant, the truncated cone then has an area  $A_W$ , an angle of aperture 2  $\Delta \theta_3$  and a generatrix  $g_W$ .

This focus factor was introduced by Liu in 2004. It takes into account the concentration of light at one point, and thus takes into account the fluence rates at a given point. This focus factor is the ratio of the two truncated cone areas  $A_{WO}$  and  $A_W$ . It is equal to:

Focus = 
$$\frac{(d_1 + d_2 + d_3)^2}{(r_1 + r_2 + r_3)\cos\theta_3 n_1} \times \left[\frac{r_1}{n_1\cos^3\theta_1} + \frac{r_2}{n_2\cos^3\theta_2} + \frac{r_3}{n_3\cos^3\theta_3}\right]$$

Then, to calculate the fluence rate, it is necessary to determine the angles of refraction  $\theta$ . Snell's law is used for this.

$$r_1 \tan \theta_1 + n_1 \sin \theta_1 \frac{r_2}{\sqrt{n_2^2 - n_1^2 \sin^2 \theta_1}} + n_1 \sin \theta_1 \frac{r_3}{\sqrt{n_3^2 - n_1^2 \sin^2 \theta_1}} = \Delta x$$

This equation can only be solved numerically, and there are different types of solvers (Reichl *et al.*, 2006).

Once  $\theta_1$  has been determined, different types of models for determining fluence rates can be applied.

There are four main models that can simulate rates of irradiation. These models are being updated continually.

#### 2.2 Multiple Point Source Summation (MPSS) model

In the MPSS model introduced by Jacob and Dranoff in the 1970s (Jacob and Dranoff, 1970), a linear UV lamp is considered as a series of discrete sources *n*, spaced regularly along the longitudinal axis of a lamp, which emits light in an identical way in all directions. The power of each discrete source is equal to  $\Phi/n$ , with  $\Phi$  being the total power of the lamp, at the wavelength in question. The fluence rate in a given volume dV is the sum of the fluence rates for each lamp element *n*, calculated for each radiant energy flux passing through the element of volume dV considered.



Figure A3: Cross section of a UV reactor. L is the length of the UV lamp considered; S is the inside radius of the reactor and s is the radius of the quartz sleeve;  $h = s \times \cot\theta$ ;  $H = h + (x - s) \cot\theta_3$ ;  $A_1$  and  $A_3$  are the transverse surfaces used to calculate the fluence rates (Bolton, 2000)

The fluence rate decreases as a function of the distance from the light source due to the dispersion and absorption of the light beam in water and in the quartz sleeve surrounding the light source.

This model can also incorporate the focus effect, in which case it is referred to as the MPSS-F model.

According to many authors, this model tends to overestimate the fluence close to the lamps (Liu *et al.*, 2004; Munoz *et al.*, 2007), and they believe that this model cannot, under any circumstances, be applied to polychromatic lamps.

#### 2.3 Multiple Segment Source Summation (MSSS) model

In the MSSS model developed by Bolton in the early 2000s (Bolton, 2000), the effects of reflection and refraction at the air-quartz-water interfaces are taken into account (essential effects that must be considered in the case of water intended for human consumption), and UV lamps are no longer considered as spherical sources, but as series of identical cylindrical segments.

In this model, the energy transmission is greater in the axis perpendicular to the surface of each element and decreases according to the cosine of the angle  $(\cos\theta_1)$  made between the perpendicular and the light source's direction of emission.

This model can also incorporate the focus effect, in which case it is referred to as the MSSS-F model.

Moreover, in this model, Bolton introduced a weighting factor to take into account the germicidal effect of polychromatic lamps.

#### 2.4Line Source Integration (LSI) model

The Line Source Integration model is the continuous or integrated version of the MPSS model. This model is mathematically identical to the point source approaches, where n, the number of point sources, is equal to infinity ( $n=\infty$ ). Nevertheless, this model has the drawback of not taking into account absorption, reflection and refraction phenomena. Liu corrected it in 2004 by adding an attenuation factor, yielding the LSI-F model.

#### 2.5Radial-Line Source Integration (RADLSI) model

Liu (Liu *et al.*, 2004; Liu, 2004) recently improved existing models by taking into account the gaps when the model was to determine fluence rates in areas close to the lamp. This RADLSI model thus combines the advantages of the MSSS and LSI models.

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# **Annex B: Radiometers**

UV reactors fitted with medium-pressure lamps must have one radiometer per lamp. Reactors fitted with low-pressure lamps must have at least one radiometer per row of lamps (US EPA 2006).

The technical characteristics of radiometers are defined below:

# 1. Measurement angle

Some radiometers have a measurement angle of 40° whereas others use 160°. In the Austrian standard ÖNORM M5973, both measurement angles are used (40° and 160°) while in the German standard DVGW W294-1 to -3, only the 40° is used. These measurement angles provide completely different measurement values. UV radiation devices tested with 40° radiometers should only be operated and monitored with 40° radiometers. Radiometers with a 160° measurement angle deliver higher values and the measurement signal reacts differently from radiometers with a smaller measurement angle.

# 2. Angular response

The angular response is a function of the angle of incidence upon the radiometer window. It is affected by the diameter of the radiometer aperture, the size of the active surface of the photodetector, the distance between the aperture and the active surface, and the effect of surfaces scattering and reflecting UV light.

An ideal radiometer has a cosine-shape response but radiometers often have a different response (Figures B1 to B4). Equipment radiometers and reference radiometers should display only the weighted irradiance in the biocidal spectral field ranging between 240 and 290 nm. Thus, it is frequently necessary to use filters that ensure a detected percentage below 10% for a wavelength > 300 nm, as recommended by the US EPA (2006).



Figure B1: Example of response from an ideal radiometer



Figure B2: Example of response from radiometers on the market



Figure B3: Radiometer with filter. Figure B4: Radiometer without filter. Percentage of intensity Percentage of intensity UV > 300 nm: 0.7 % of the totaIUV > 300 nm: 41 % of the total

# 3. Measurement windows

Measurement windows must be made of UV-resistant, stable material (for example: stainless steel, PTFE). Quartz-glass windows must have a transmission spectrum such that transmission above 250 nm is greater than or equal to 90%.

Devices fitted to medium-pressure reactors must also have transmission greater than or equal to 85% in the spectral field between 200 and 250 nm.

The German standard DVGW W294-2 specifies that radiation above 300 nm must not contribute to more than 1% of the signal and radiation below 240 nm, not to exceed 5% (DVGW, 2006).

# 4. Linearity

Linearity must range from 0.1 W/m<sup>2</sup> to 250 W/m<sup>2</sup> for radiation of a wavelength of 254 nm.

The measurement range of the control radiometer must include the range of admissible irradiances. The range between E1 and E2 must be readable in 3% increments of the range.

E1: lowest irradiance value in the operational field in W/m<sup>2</sup>

E2: highest irradiance value in the operational field in W/m<sup>2</sup>

A: smallest irradiance difference still readable on the radiometer control display (W/m<sup>2</sup>).

The functioning of the radiometers must be verified regularly. The value displayed by a radiometer placed on the equipment must be checked by comparing its measurement with that of a reference radiometer. For devices with flow rates over  $100 \text{ m}^3/\text{h}$ , the German DVGW standard recommends checking it monthly. For other devices, checking every six months is recommended. The measured value obtained with the operating UV radiation radiometer must not exceed the measured value obtained with the reference radiometer by more than 5%. If the displayed value differs by more than 10% from the value measured by the reference radiometer, the display should be aligned. After 10,000 hours of use, or if the difference is more than 20% (which is already an excessive value) and no later than two years after the radiometer has been commissioned, calibration is required.

# 5. Reference radiometers

Requirements for reference radiometers are prescribed in the ÖNORM M 5873-1:

- They must be standardised by an official authorised body; their measurement range must be between 0.1 W/m<sup>2</sup> and 250 W/m<sup>2</sup> at a wavelength of 253.7 nm.
- Their uncertainty must be less than 10%.

#### **References Annex B**

- **DVGW, 2006a.** Technical standard W294-1 Appareil de désinfection UV dans l'approvisionnement en eau Partie 1 : exigences relatives à la qualité, au fonctionnement et à l'exploitation.
- **DVGW, 2006b.** Technical standard W294-2 appareil de désinfection UV dans l'approvisionnement en eau Partie 2 : examen de la qualité, du fonctionnement et de l'efficacité de désinfection.
- **DVGW, 2006c.** Technical standard W294-3 appareil de désinfection UV dans l'approvisionnement en eau Partie 3 : Fenêtres de mesure et capteurs pour la surveillance radiométrique d'appareils de désinfection UV - exigences, examen et étalonnage.
- **ÖNORM, 2001.** Standard ÖNORM M5873-1 Plants for the disinfection of water using ultraviolet radiation requirements and testing part 1: Low pressure mercury lamp plants.
- **ÖNORM, 2003.** Standard ÖNORM M 5873-2 Plants for the disinfection of water using ultraviolet radiation requirements and testing part 2: medium pressure mercury lamp plants. 40 p.
- **US EPA, 2006.** UVGM-Ultraviolet disinfection guidance manual for the final long term 2 enhanced surface water treatment rule.

# Annex C: Photorepair and dark-repair mechanisms

# 1. Photoreactivation

Photoreactivation enables microorganisms considered temporarily inactivated by UV radiation to regain their metabolic activity and/or culturability by the action of specific enzymes, the photolyases.

This family is composed of three groups (Todo, 1999):

- Cyclobutane pyrimidine dimer photolyases (CPD-photolyases), which are themselves divided into two categories: class I and class II CPD-photolyases, according to the sequence of the amino acids of which they are made. Class I photolyases cover all microorganisms whereas those of class II mainly concern plants and animals (Weber, 2005);
- (6-4) photolyases, found only in some eukaryotic species (Moreno de Lima-Bessa *et al.*, 2008); and
- Cryptochromes (CRY), found only in plants, animals and humans.

These DNA photolyases are soluble proteins with a molecular weight of between 55,000 and 65,000 daltons, identified in a variety of organisms ranging from bacteria to multicellular eukaryotes. All photolyases contain at least one chromophore, which is flavin adenine dinucleotide (FAD) and the majority of them contain a second chromophore, which is either methenyltetrahydrofolate (MTHF) (as in *Escherichia coli*), 8-hydroxy-5-deazariboflavin (8-HDF) (Byrdin *et al.*, 2004), or flavin mononucleotide.

FAD has photochemical activity and plays a direct role in the cleavage of dimers. MTHF and 8-HDF are auxiliary photon sensors which transfer absorbed energy to FAD. Photolyases that have both chromophores (FAD + auxiliary chromophore) are the most effective for cleaving dimers, and therefore in repairing DNA damage (Beukers *et al.*, 2008).

To repair the inactivated DNA, photolyases must have the FAD cofactor in its fully reduced form (FADH<sup>-</sup>). The presence of the second chromophore is not necessary for observing DNA repair; its only function is to increase the absorption of light in the near-UV and visible regions where FADH<sup>-</sup> absorbs only low levels (Mu *et al.*, 2005), and to transmit the energy gained to FADH<sup>-</sup> to yield its excited singlet form <sup>\*</sup>FADH<sup>-</sup>. This electron transfer is very fast: 134 picoseconds (ps) in the case of MTHF and 50 ps in the case of 8-HDF. If there is no second chromophore, <sup>\*</sup>FADH<sup>-</sup> can also be generated directly by photo-excitation of FADH<sup>-</sup> (Weber, 2005).



Figure C1: Repair mechanism of Cyclobutane Pyrimidine Dimers (CPDs) by CPD-photolyase (Weber, 2005).

Once formed, FADH transfers an electron to the cyclobutane dimers to generate one flavin radical semiquinone (FADH) and one CPD radical ion, in a very short time (approximately one hundred picoseconds). The bonds C(5)-C(5'), then the bonds C(6)-C(6'), contained in this radical anion are then broken, and the two original pyrimidines are then regenerated; whereas FADH is reactivated in the form FADH.



Figure C2: Repair mechanism of (6-4) photoproducts by (6-4)-photolyase (Weber, 2005).

The repair capacity of microorganisms is known to differ significantly depending on the species and strains concerned.

# 2. Dark-repair

There are three different types of light-independent repair involving the use of various enzymes, but all follow the same mechanism as shown in Figure C3:



Figure C3: Excision repair mechanisms (Prescott et al., 2003). Repair by excision of a thymine dimer that causes deformation of the double helix. Repair endonuclease (UvrABC endonuclease) is coded by the genes UvrA, B and C.

Repair by recombination is an important mechanism because it avoids blocking replication for hours while awaiting elimination of lesions by excision-resynthesis. Its role is particularly important in that it allows the repair of damage not detectable by excision-resynthesis mechanisms, such as alterations that block DNA synthesis without causing distortion of the helix. The phenomenon of repair by recombination has been demonstrated in bacteria, however, its existence in eukaryotic cells is uncertain.

The double mutant strains uvr<sup>-</sup> recA<sup>-</sup> of *E. coli* that are deficient both in the resynthesis excision system and in repair by recombination, lose all of their repair ability and cannot remove most of the lesions. In these cells, attempts at replication produce fragments of DNA whose size corresponds to the expected distance between two dimers of thymine. Thus, it is shown that dimers are a lethal obstacle to replication when the RecA function is impaired and explains why a double mutant cannot tolerate more than one or two dimers in its genome, whereas a wild-type cell can overcome the presence of around fifty dimers.

The RecA protein intervenes by forming a protective filament around the single-strand DNA from the gap due to replication system jumps. The complex single-stranded DNA - RecA protein is a substrate that can match the single strand with a homologous strand, the first step of induced recombination. The RecA protein thus promotes apposition between DNA strands that will be used to restore the double helix.

The recA<sup>-</sup> mutants are almost completely impaired in both genetic recombination and repair response. Indeed, The RecA protein plays a critical role in these two mechanisms, where it performs the function of:

- exchanging strands between DNA molecules, a central activity in recombination,
- exchanging single strands between newly synthesised molecules, involved in repair by recombination.

Mutations in the *recA* gene and in other *rec* genes have identified pathways of repair. Some components involved in the repair are also involved in recombination, but they are not exactly the same since some mutations affect one mechanism, but not the other.

The properties of *recA recX, recA recY and recX recY* double mutants (here, *recX and recY* means two different *rec* genes) suggest that *recA* intervenes in at least two different pathways. One also involves the *recB* and *recC* genes, which encode two subunits of exonuclease V, whose action is controlled by other components of this pathway, including the RecA protein itself. The *recF* gene, whose function is unknown, identifies another pathway for recombination and repair by recombination. In fact, despite the undeniable progress of research on these mechanisms, molecular details of the systems of repair by recombination are still not precisely known.

#### **References Annex C**

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# Annex D: Normative and regulatory context

# 1. Regulations of Member States of the European Union

## 1.1 Germany

WIHC regulations are outlined in a document entitled "*Trinkwasserverordnung*" [Drinking water regulation] of 21 May 2001 (UBA, 2001). An article in this text (§ 11) stipulates that a list of approved products and systems for disinfection be published by the Federal Ministry of Health. This list was updated in December 2009 (UBA, 2009) and the UV radiation systems are described in Part II of this list for use in disinfection at wavelengths of between 240 and 290 nm. The list specifies the applicable technical rules, which are technical specifications DVGW W294-1, W294-2 and W294-3 and establishes the following requirements:

- only UV radiation equipment with a proven disinfection efficacy of at least 400 J/m<sup>2</sup> (according to the DVGW W294-2 standard) are authorised;
- operational characteristics (maximum flow rate and minimum corresponding radiation power) specified for the device in the test report as well as in the DVGW certificate must be observed during use of the reactor.

The notes state that it is possible to continue using non-certified UV disinfection equipment only up to 30 June 2012 if their disinfection efficacy has been demonstrated in an individual test or in small private water production facilities with the approval of the competent authorities. The notes also specify that these systems are not applicable for maintaining disinfection capacity in the distribution system.

In addition, the list specifies that when disinfecting surface water or water influenced by surface water, it is necessary to remove as many particles as possible prior to disinfection and to obtain turbidity values in the range of 0.1 to 0.2 FNU, and below if possible, before passage through UV radiation.

## 1.2 Austria

The regulation for WIHC is described in the Austrian food book, Chapter B1 "Drinking water" in paragraph 4.5, which refers to UV radiation as a reliable disinfection system. It states that disinfection should be verified by the absence, in 250 mL, of *E. coli* and coliform bacteria, Enterococci, *Pseudomonas aeruginosa, Clostridium perfringens*, total bacteria at 22°C and 37°C. In addition, in paragraph 4.15 of Chapter B1 the book outlines requirements for the use of this technology, especially concerning application of the dose of 400 J/m<sup>2</sup>, wavelength of 253.7 nm and certification of the device, relying on Austrian standards in force and states:

- based on national standards establishing all the requirements for the safety of disinfection by UV radiation, test results on full-scale systems of each type of reactor are specifically requested;
- as part of validation of a UV reactor, a biodosimetry test is required to help define the operating parameters of each reactor so that it delivers a dose equivalent to at least 400 J/m<sup>2</sup> (water flow (m<sup>3</sup>/h, water transmittance (%), UV irradiance (W/m<sup>2</sup>)). These data are specified in the certificate issued by the Austrian Association for Gas and Water (OVGW);
- the operating parameters must be recorded in the water production plants and must match those specified in the certificate. Alarms are defined at each field site.

Austria also published an official statement summarising the changes in requirements since 1983 on UV radiation technology, which stipulates the procedures for managing "old facilities", concluding that these facilities could not guarantee adequate disinfection and stressing the urgency of replacing them (*Bundesministerium für Gesundheit, Familie und Jugend & Trinkwasser* [Austrian Federal Ministry of Health, Family and Youth & Drinking water], 2007).

# 1.3 United Kingdom

WIHC is regulated by a document applicable to England and Wales entitled "The Water Supply (Water Quality) Regulation 2001" with various amendments. Part VII of this document includes Regulations 25 to 33 concerning water treatment. Based on this, an authorisation system for treatment products and systems in contact with WIHC was put in place and specific guidelines for the use of UV reactors were published in February 2010 by the Drinking Water Inspectorate (DWI). These guidelines reiterate that the maximum limit of turbidity for disinfection is 1 FNU. Biocides added to maintain a residual disinfectant concentration in the water distribution pipes must be added after the UV radiation disinfection step.

Key points are to be verified for validation of the equipment, especially comprehensive biodosimetric testing. Reactors must operate within the limits defined in the tests. Minimum monitoring requirements are also defined, especially for water flow, the condition of the lamps, the UV dose, turbidity and lamp replacement.

Requirements	Conditions		
For conducting the tests:	<ul> <li>Flow rate</li> <li>UV dose (J/m<sup>2</sup> measured by UV radiometers)</li> <li>Status of the lamps</li> </ul>		
For conducting biodosimetry tests:	<ul> <li>Testing on the actual reactor size, or a reactor exactly like the one to be used</li> <li>Inactivation of a test microorganism whose behaviour is well known in terms of the dose of UV radiation, i.e., that it was measured properly and is representative of claims in relation to its intended uses</li> </ul>		
To determine during the validation tests:	<ul> <li>Transmittance and absorbance of the water</li> <li>Aging of the lamps and presence of deposits on the sleeves</li> <li>Measurement uncertainty of operating radiometers</li> <li>Distribution of the dose of UV radiation resulting from different velocity profiles through the reactor to demonstrate that at every point of the reactor the water receives the minimum dose required</li> <li>Failure of the lamps or other critical component of the reactor</li> <li>Configuration of the piping at the reactor inlet and outlet</li> </ul>		

Table DIV: Minimum requirements for validation testing (DWI, 2010)

Recommendations for monitoring and verifying the system at the plant are specified. Concerning the formation of by-products, the conventional indicators used are not pertinent to this type of disinfection but the products of photolytic reactions, such as nitrites may appear. The materials used in the reactors must meet regulatory requirements and have proof of compliance with the health requirements issued by the DWI. In the event of lamp breakage, the introduction of mercury into the WIHC and a list of risk factors for rupture are included in the guidelines.

## 1.4 Ireland

There are no national regulations for authorising treatments applicable to WIHC but general requirements are specified in Paragraph 13 of the drinking water regulation. However, competent services are instructed to use the products and systems listed in the United Kingdom or an equivalent European system.

Ireland requires that UV radiation treatment systems, which have increased in number in the territory in recent years, meet the following conditions:

- 1. Validation certificate, usually based on American or German standards for lamps, indicating range limits (lamps output/power);
- 2. Confirmation that the system has a operating lamp alarm in order to confirm that it remains continuously within the range of operation validated by the above certificate;
- 3. Verification from recordings to ensure that the system operates in the range provided for;
- 4. Confirmation that an automatic backup system is provided in case of malfunction, so nondisinfected water is not sent through the system;
- 5. Details on methods the producer uses to ensure that water is not contaminated in the pipelines, since there is no residue with this disinfection system.

In addition, two control requirements are applied:

- 1. If treatment with UV radiation is required to solve a problem with the quality of the resource, the action plan must correspond to the recommendations of the US EPA, in which case the administration makes sure that the criteria are met before authorising UV as a solution.
- 2. During an audit of a water production chain including a UV radiation disinfection system, the administration verifies that all requirements have been met and if not, the producer must commit to solutions to meet the conditions required.

#### 1.5 Sweden

While there is a positive list of water treatment products, there is none for systems. General requirements are indicated in the ordinance on WIHC (*LIVSMEDELSVERKET* [Swedish National Food Administration], 2006) which specifies that:

- Disinfection equipment must be fitted with warning systems in case of malfunction;
- The distribution manager must make sure that disinfection is carried out properly.

#### 1.6 Portugal

A system of authorisation for the products and systems used for treatment of WIHC is being developed and a committee was created to conduct the evaluations. UV reactors are still not in common use.

#### 1.7 Italy

Management of WIHC is governed by a national decree in accordance with European Directive 98/83/EC. Its Article 9 reports the general principle regarding the safety of treatments used. However, the country states that it is developing stricter rules to assess the risks associated with treatment products and systems used for the production of WIHC. UV reactors for water disinfection do not appear to be in widespread use in Italy.

## 1.8 Cyprus

Quality control of the supply of WIHC is considered to be adequate and there is no national regulation to authorise treatment products and systems for WIHC. The regulatory document specifying the limits deriving from the European Directive 98/83/EC is entitled "The quality of water intended for human consumption (monitoring and control)" Law 87(I)/2001. UV radiation disinfection is not used to treat water in Cyprus.

#### 1.9 Belgium

Water treatment products are regulated and subject to a positive list, but systems are not listed. There is no specific regulatory provision applicable to UV reactors used for water disinfection.

## 1.10 Slovenia

There is no specific regulation for water treatment systems. Only products are listed and compliance with microbiological limits of the water supply is verified through monitoring and inspection.

# 2. Other regulations

### 2.1 World Health Organization (WHO)

The guidelines on the quality of WIHC published by the WHO (WHO, 2008) give the doses of UV radiation for inactivation of various microorganisms (Table DV).

Table DV: Percentages of inactivation obtained by UV irradiation for various microorganisms (WHO, 2008)

Microorganism	Inactivation
Bacteria	2-log for 70 J/m <sup>2</sup>
Viruses	2-log for 590 J/m <sup>2</sup>
Protozoa	
Giardia	2-log for 50 J/m <sup>2</sup>
Cryptosporidium	3-log for 100 J/m <sup>2</sup>

In the WHO document on water treatment and control of pathogens, (WHO, 2006), Section 3 is dedicated to disinfection and Section 3.3.5 is devoted to UV radiation and includes the data presented in Table DVI.
Microorganism	Dose for 4-log inactivation (J/m²)	Water source
Bacteria		
Bacillus subtilis spores	310	Laboratory
Escherichia coli	200	Laboratory
Salmonella typhi	300	Laboratory
Vibrio cholerae	6.5	Laboratory
Viruses		
MS2 phage	500	1 groundwater
	640-930	11 groundwaters
	1000	Laboratory
Coxsackie A	300	Laboratory
Hepatitis A	60-150	3 groundwaters
	160	Laboratory
Poliovirus	230-290	8 groundwaters
	300	Laboratory
Rotavirus- Wa	500	Laboratory
Rotavirus SA11	400	Drinking water
Adenovirus	1860	4 laboratory studies

Table DVI: UV doses needed to obtain 4-log reductions in various microorganisms (WHO, 2006)

# 2.2 Norway

On its English website, the Norwegian Institute of Public Health (NIPH) indicates that disinfection of water by UV reactors is fairly common in Norway, especially in small water distribution systems. Reactors using low and medium-pressure lamps are recognised. Since the 1970s, all approved systems have been evaluated by the NIPH to ensure that they comply with standard conditions and the list of approved reactors is available on the website<sup>2</sup>. A dose above 400 J/m<sup>2</sup> obtained by biodosimetric testing is required before a device can be placed on the market. German, Austrian and US EPA protocols are accepted, the approved test centres are those recognised for these three standards, since the country itself does not have a national testing centre. The recognised efficacy of reduction is 6-log for bacteria that are hazardous to health in WIHC, 4-log for spores of bacteria of interest and 2-log for viruses of interest, provided that the operating conditions for delivering the required dose are met.

# 2.3 Switzerland

The decisive parameter for the performance of disinfection is the UV dose. In this regard, the *Société Suisse de l'Industrie du Gaz et des Eaux* [Swiss Society for the Gas and Water Industry] (SSIGE) established a Regulation, SSIGE W/TPW 152 (SVGW/SSIGE, 2008), in September 1998, which

<sup>&</sup>lt;sup>2</sup> http://www.fhi.no/

stipulates that WIHC follow the German and Austrian standards relating to water disinfection systems using UV radiation.

### 2.4 New Zealand

The New Zealand Ministry of Health has published three papers concerning disinfection of WIHC by UV irradiation. It does not recognise this treatment for virus disinfection.

- 1. Acceptable limits in WIHC (Ministry of Health, 2005) that include, in Paragraph 5.16, mandatory efficacy criteria for *Cryptosporidium* for UV reactors. Compliance with German or Austrian standards or with Part 1 of the American standard for a dose of 400 J/m<sup>2</sup> is recognised. If the American approach (other than Part 1) is followed, in order to be approved, low-pressure lamps must demonstrate compliance for a RED of 360 J/m<sup>2</sup> and medium-pressure lamps for a RED of 420 J/m<sup>2</sup>. This document also outlines obligations for the use of reactors, including obligatory preliminary filtration, turbidity at intake of at least 2.0 FNU for a maximum of three minutes and less than 5% of water flow greater than 1.0 FNU (under continuous monitoring); a UV transmittance of at least 80% cm<sup>-1</sup>; in the minimum flow rate. This document also provides the points to be verified by the water producer deploying the reactor; finally, it has self-inspection obligations depending on the size of the population served.
- 2. The water quality management guidelines (Ministry of Health, 2005) include, in Section 15 on treatment systems used for the disinfection of water, Subsection 15.5.5 on UV radiation. The document emphasises the absence of listed hazardous by-products at the treatment doses implemented and the need for characterising the effects of treatment on organic matter and any possible health effects that may be involved. It states that research is always needed even if the system is known for its efficacy, including on the phenomena of photorepair. This document also underlines the difficulty of validating a reactor given the many cumulative uncertainties (distribution of water flow in the reactor, response of the radiometer depending on the angle of the light, the different qualities of water and variations in lamp power). The document specifies that the lamp supplier must indicate for how many hours it can operate, at what power, and must apply a reduction on this power after approximately 100 hours of operation and a maximum number of times it can be switched on per day. It shows the quality limits on water intake in detail and specifically dictates that there must be no trace of permanganate and ozone. The document also provides details for the design of the reactors, their installation, monitoring and use.
- 3. The Public Health Risk Management Plan Guide of the Ministry of Health (Ministry of Health, 2001) includes a sheet on the treatment systems dedicated to disinfection by UV irradiation. This document lists the causes, preventive measures, and means of control using these preventive measures: the parameters to be monitored and limits requiring a corrective action and finally, the corrective actions to implement. This table identifies six high and medium level causes.

### 2.5 USA

The "Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR)" of January 2006 (US EPA, 2006) reinforced control of pathogenic microorganisms in WIHC. It acknowledges UV radiation as a disinfection technique and grants it a reduction credit based on the delivered dose, the reactor's validation tests and conditions for its implementation. The accepted reductions are shown in Table DVII.

Reduction credit in log	<i>Cryptosporidium</i> UV dose in J/m²	<i>Giardia lamblia</i> UV dose in J/m²	Viruses UV dose in J/m²	
0.5	16	15	390	
1.0	25	21	580	
1.5	39	30	790	
2.0	58	52	1000	
2.5	85	77	1210	
3.0	120	110	1430	
3.5	150	150	1630	
4.0	220	220	1860	

Table DVII: reduction credit and UV dose required for Cryptosporidium, Giardia lamblia and viruses (US EPA, 2006)

Section 141.716 of the LT2ESWTR outlines the requirements for use of a UV reactor, including the biocidal wavelength, fouling of the lamps, uncertainty of UV radiometers, velocity profiles in the reactor, etc. Conditions for validation are described in this text along with follow-up procedures for self-monitoring by the water producer. It specifically requires that 95% of the water treated on a monthly basis be produced under conditions that ensure proper compliance with the required dose.

The U.S. Environmental Protection Agency generated various documents in April 1999, including a guide to alternative disinfectants and oxidants (US EPA, 1999). Its Section 8 is dedicated to UV radiation and details the components of a reactor, its design and the parameters to be taken into account for the transmittance of water. This document also includes data on the efficacy of UV radiation on the maintenance of reactors. It indicates that the treatment is incompatible with water that has high iron, calcium, turbidity and phenol concentrations, and states that it should be applied just before entering the distribution system. The Agency has also published guidelines for the validation of UV reactors within the framework of the LT2ESWTR in 2006.

In 2003 the National Water Research Institute (NWRI) also published Ultraviolet Disinfection Guidelines for Drinking Water and Water Reuse (Second Edition) (NWRI-AWWARF, 2003).

The country has an NSF/ANSI (National Science Foundation/American National Standards Institute) international and national standard for ultraviolet microbiological water treatment systems NSF/ANSI 55 – 2007 (NSF/ANSI, 2007).

### 2.6 Quebec

Quebec has a regularly updated procedure for analysing WIHC treatment technologies. This document states that any proposed facility or modification of a WIHC production facility supplying more than 20 people must be authorised by the Ministry of Sustainable Development, Environment and Parks (MDDEP) pursuant to the Law of Environmental Quality. A guide to the design of production facilities and WIHC distribution systems is also available to assist stakeholders with the authorisation system.

 The document detailing the authorisation procedure (MDDEP, 2008) specifies that the Quebec Drinking Water Treatment Technologies Committee (CTTEP) is in charge of reviewing the authorisation application dossiers and issues technical evaluation sheets. As a prerequisite to an application for authorisation, the treatment technology must meet the regulatory standards, or recommendations when there is no standard, for drinking water quality. In order to be authorised, a system must satisfy four levels of development: experimental, pilot-scale, full-scale, and tested, each with objectives, requirements on the discharge of treated water, assessment criteria, and for the full-scale and tested levels, a requirement for MDDEP authorisation for a proposal and a limited number of full-scale facilities. The sheets specify the critical parameters for water before treatment. Annex B of this document refers in part to UV reactors and requires validation by an accepted biodosimetric method to confirm the effective dose provided by a reactor under different operating conditions. The manufacturer must provide the results of these tests and specify the protocol used and the independent body that supervised the tests. The protocols referenced are German (DVGW-W294), Austrian (ÖNORM M 5873-1) and American (NWRI-AWWARF and NSF 55). The 2003 and 2006 versions of the US EPA protocol (UVGM) may be accepted.

2. The guide to design of facilities includes Section 10 (MDDEP, 2006) related to disinfection and control of by-products, which specifies the mandatory minimum reduction to obtain, in various types of raw water and depending on the target organisms (Cryptosporidium, Giardia and viruses). There are also additional objectives for surface water (or water influenced by surface water) based on their total coliform concentrations. This document provides a table comparing different methods of disinfection, and UV radiation treatments are shown as having an excellent efficacy against Cryptosporidium, very good efficacy for Giardia and acceptable efficacy against viruses. Subsection 10.4.5 is devoted to UV radiation. It states that the system must be applied at the point in the systeming chain where the UV transmittance of the water is the highest or where fouling of the lamps is kept to a minimum. It also details the doses designed for the reactor, namely the doses, depending on the type of water, for achieving a 3log reduction in protozoa and 2- or 4-log in viruses; the required doses range from 400 J/m<sup>2</sup> to 1200 J/m<sup>2</sup>. It indicates that to achieve 800 J/m<sup>2</sup> for example, the designer uses two reactors validated at 400 J/m<sup>2</sup> in series or deduces 25% from the maximum flow allowed for a reactor validated at 600 J/m<sup>2</sup>. This document also indicates the monitoring and alarm systems with which the reactors must be fitted. Concerning fouling of the lamps, it specifies that the water being treated must not exceed the following limits: 0.3 mg/L for iron, 120 mg/L for hardness and 0.05 mg/L for manganese.

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# Annex E: Emerging technologies

# 1. Lamps

## 1.1Light-Emitting diodes (LEDs)

#### Design and emission spectrum

Light-Emitting Diodes (LEDs) are semiconductor components which, when crossed by a current, emit light, with an energy generally proportional to the energy current.

This light emission is due to the radiative recombination of electron-hole pairs in type p-n heterojunctions (Figure E1).

The first diodes developed in the 1960s were made of an alloy of Ga/Al/As (aluminium gallium arsenide) and GaAs (gallium arsenide), with characteristics that limited the available colours to red and infrared. It was only in the 1990s that new nitride-based materials appeared (GaN, AlGaN, InGaN) giving access to shorter wavelengths, such as green, blue, and up to near ultraviolet.



Figure E1: Heterojunctions within LEDs enable various wavelengths of radiation to be obtained

LEDs are the basis of so-called Lambertian emitters, meaning that light is emitted equally in all directions. One of the differences in the use of LED lighting compared with more traditional sources is that the elementary power of each source is low, thus the power is obtained by multiplying the elementary sources. One result, related to the laws of optics, is that to form a directional beam, each source must have its own optical components (flat, conical, dome-shaped, bullet-shaped lens, etc.). This optical system can be cast directly on the diode or added in front of the diode.



Figure E2: Examples of LEDs with dome lens and flat lens (www.biouvled.com)

LEDs have only recently been developed in the near UV-C range (250 nm). Thus a whole range of LEDs operating between 255 and 405 nm are on the market. Due to the nature of the metals they contain, they are all monochromatic.



Figure E3: Spectrum of LEDs currently on the market

#### **Technical characteristics of LEDs**

LEDs have physical characteristics that make them potential sources of UV radiation for water disinfection.

First, they have a much longer lifetime than conventional mercury vapor lamps (minimum 100,000 hours as opposed to 10,000 hours for low-pressure UV lamps) (Crawford *et al.*, 2005).

They contain no toxic substances such as, for example, the mercury in conventional UV radiation lamps. They consume less electrical energy and yield large amounts of electric power/light output. In fact, they do not emit any heat: the electrical power is totally dedicated to light radiation (Vilhunen and Sillanpää, 2009).

Their small size (between 10 and 15 cm in length, and between 2 and 10 cm in width), the nature of the materials forming the optical system (doped glass versus quartz in mercury vapor lamps), and the ability to use only the wavelengths required make them UV radiation sources that can serve as alternatives to conventional mercury vapor lamps. Indeed, they are less fragile than conventional UV radiation lamps and their small size makes it possible to devise new reactor designs, ensuring optimised contact between the UV radiation and the flow of water to be treated by adjusting the number of LEDs and their wavelengths.

However, to design a reactor for treating large flows, many LEDs will be needed, and their cost is still very high currently. This explains why there are still no LED UV reactors on the market for disinfection in drinking water purification plants. Note, however, that a sewage plant in Hong-Kong has been using an LED UV reactor in tertiary treatment since 2003 (Close *et al.*, 2006).

Current applications thus involve treating only small volumes or low flow rates (micro and nanoelectronics, biological and medical laboratories, etc.).

A study by Sensor Electronic Technology Inc. (SETI) (Reed *et al.*, 2008) shows that the cost of LEDs continues to decline, and estimates that from 2014 onwards, the cost of LEDs at 255 nm should reach the current prices of LEDs at 365 nm.



Figure E4: Price trends of LEDs (Reed et al., 2008)

#### **Disinfection efficacy**

There are very few scientific articles (only four) focusing on the use of LEDs for water disinfection.

Hamamoto *et al.*, 2007, used a high-energy UV LED (up to 504 J/cm<sup>2</sup>) emitting at a wavelength of 365 nm and tested its efficacy on five bacteria: *Escherichia coli* DH5α, Enteropathogenic *Escherichia coli* (EPEC), *Vibrio parahaemolyticus*, *Staphylococcus aureus* and *Salmonella enterica*. Reductions reached 5.7-log for the first one in 75 minutes (at 315 J/cm<sup>2</sup>), 5.2-log for the following three in 60 minutes (at 252 J/cm<sup>2</sup>) and 3.4-log for the last one (at 504 J/cm<sup>2</sup>). The same authors, in another publication (Mori *et al.*, 2007), also showed the effect of temperature and pH on disinfection efficacy. It appears that the optimum temperature and pH are respectively 20°C and pH 8.

The same authors (Mori *et al.*, 2006) used these same LEDs at 365 nm (15 mWs/cm<sup>2</sup>, which is 150 J/m<sup>2</sup>), in static mode, but by using a device with eight LEDs in series. The results showed a germicidal efficacy of 100% for *Escherichia coli* (in 30 minutes) and for *Vibrio parahaemolyticus* (in 10 minutes). However, the initial number of bacteria was not stated in this publication.

Vilhunen and Sillanpää, 2009, studied the efficacy of LEDs emitting at wavelengths of 269 and 276 nm for the inactivation of *Escherichia coli*. Testing was conducted in static mode with an initial bacteria level of 10<sup>7</sup> CFU/cm<sup>3</sup>. After 25 minutes, it was shown that the reduction was 7-log, for both wavelengths. Efficacy was reduced when the medium was turbid: (4-log reduction in 25 minutes for a turbidity of 1.45 nephelometric turbidity units (NTU).

In contrast, there are more than twenty international patents which relate to the use of LEDs for water disinfection. These patents cover both mobile disinfection systems (Maiden, 2003, 2006; Levy, 2006) and reactors that can process high volumes and flow rates (Schlesser and Lemunyon, 2009; Harbers, 2006; Knight *et al.*, 2009) but there are no serious scientific data supporting these patents.

### 1.2Hollow cathode lamps

#### Design and emission spectrum

Hollow cathode lamps are discharge lamps where the cathode is the covering (Figure E5). The authors (Soloshenko *et al.*, 2006) used various gases (air, oxygen, water vapor, deuterium-oxygen mixture) at low pressure. They compared the emission spectra of each of these gases or mixture with that of a conventional mercury vapor lamp (Figure E6). Except for the air lamp (a), which has several maximum wavelengths between 215 and 270 nm, the other gases emit mainly in the far UV at around 220 nm.



Figure E5: Diagram of the experimental device.

(Soloshenko et al., 2006).

- Cathode envelope 1-
- 2-Anode

- KU-1 quartz window KU-1 3-
- Ku-1 quartz Petri dish Plasma discharge 4-
- 5-



Figure E6: Emission spectra of hollow cathode lamps, containing various gases

(NB: the irradiation intensities were standardised)

(Soloshenko et al., 2006)

- (a) Air at pressure of 0.2 Torr
- (b) Oxygen (0.1 Torr)
- (c) Water vapor (0.1 Torr)
- (d) Deuterium (0.35 Torr) mixed with oxygen (0.08 Torr)
- (e) Mercury vapor lamp

#### **Disinfection efficacy**

Hollow cathode lamps are more effective against *E. Coli* than mercury vapor lamps (Figure E7) because of the range of wavelengths generated.



Figure E7: Survival curves during irradiation of an aqueous suspension of 2 10<sup>6</sup> CFU/mL of Escherichia coli.

Reduction rate of E. Coli based on the dose of exposure to UV radiation emitted by DB-30- and PRK-400-type mercury vapor lamps and by hollow cathode discharge (HCD) radiation in air, oxygen, water vapor and a mixture of deuterium and oxygen. (Soloshenko et al., 2006)

### 1.3Discharge lamps

#### Design and emission spectrum

Excimer-type discharge lamps are composed of an inner electrode and an electrode located on the exterior of a quartz cover. This exterior electrode is perforated to allow the passage of UV radiation (Figure E8).



Figure E8: Cylindrical configuration of dialectric barrier discharge excimer lamp (From Naunovic, 2008)

Several gases or gas mixtures can be used. Nanovic *et al.* (2008) worked with an Xe – Br mixture emitting in the vicinity of 280 nm.

Figure E9 shows the emission spectra of various noble gases and mixtures with halogens (Sosnin *et al.*, 2006). This author compares the effect of a barrier discharge (B-A) and a capacitor discharge (D-A).



Figure E9: Emission spectra of various pure noble gases and mixtures of noble gases with halogens (Sosnin et al., 2006)

#### **Disinfection efficacy**

Discharge lamps show a relatively low efficacy for the inactivation of spores of *B. subtilis*. To improve the irradiation efficacy, the authors of the study mounted a spiral baffle (Figure E10) around the lamp. However, its efficacy remains barely satisfactory at a low flow rate and declines quickly when the flow increases (Figure E11).



Figure E10: Example of a reactor fitted with a spiral baffle (from Naunovic et al., 2008)



Figure E11: Comparison between measured inactivation and inactivation calculated by numerical simulation of B. subtilis spores for a reactor fitted with excimer lamps and a Tw of 99% (Naunovic et al., 2008)

#### 1.4Conclusion about lamps

Studies seeking to replace mercury vapor lamps (which can cause pollution) by other types of UV radiation sources, show some efficacy, especially with LEDs, which emit at wavelengths of 365 nm, or with discharge lamps emitting at a wavelength of 280 nm. Hollow cathode lamps are effective but their emission spectrum focused on wavelengths of 210 nm may raise concerns about the formation of by-products. The studies published to date do not address the chemical consequences of irradiation.

# 2. New configurations

In addition to the studies cited above which seek to replace mercury vapor lamps, some authors have proposed new configurations to improve the efficiency of irradiation.

### 2.1 Optical fibres

A system using optical fibres to deliver UV radiation from the lamp to the reactor can offer a real improvement in reactor performance.

Lu studied the use of optical fibres to 'carry' the radiation from the mercury vapor lamp to the reactor (Lu *et al.*, 2008). The system consists in focusing the radiation emitted on the end of a cluster of fibres that deliver the radiation throughout the contact chamber (Figure E12).



Figure E12: Example of an experimental device fitted with optical fibres (Lu et al., 2008)

The study by Lu *et al.* shows that the efficacy of *E. coli* inactivation is based on the number of fibres used (Figure E13).



Figure E1: Curves for inactivation of E. coli by exposure to UV as a function of irradiation time and the number of optical fibres used (Lu et al., 2008).

Of greater interest is the part of the study concerning the effect of turbidity, iron and humic acids (Figure E14). However, the study made no comparison with a conventional reactor.



Figure E14: Curves for inactivation of E. coli by exposure to UV as a function of irradiation time and the turbidity of the water (left), of the presence of dissolved iron (centre) and the presence of humic acids (right).

### 2.2HOD<sup>®</sup> system

This technique uses medium-pressure high-intensity UV lamps that are mounted on the exterior of the reactor (Figure E15). The reactor is composed of a quartz sleeve fitted with one or two lenses, depending on whether the device has one or two lamps. UV radiation is, in part, reflected towards the

radiation chamber via a mirror. According to the manufacturer, this helps to optimise distribution of the irradiation dose in the reactor.



Figure E15: Diagram of the HOD system

The lenses are covered, lamp side, with an absorbant sleeve which greatly attenuates radiation at wavelengths below 240 nm and thus significantly reduces the risks of formation of undesirable by-products.

Tests performed using commercial devices showed good disinfection efficacy with doses close to 400  $J/m^2$  (Table EI).

Microorganisms	Transmittance UV at λ=254 nm (%)	RED (J/m²)	Mean concentration (4 replicas per series) (unit log – N log)		Observed log reduction	
			Reactor inlet	Reactor outlet	Mean	Standard deviation
MS2 phages	94.5	480	4.06	1.42	2.6	0.1
Enterococcus Faecalis	89.9	400	6.74	0	6.7 minimum	0.7
Pseudomonas aeruginosa	90.1	400	7.32	0	7.3 minimum	0.5
Spores of Clostridium bifermentans	93.3	400	5.70	2.88	2,8	0.6
Escherichia coli	93.1	400	7.56	0	7.6 minimum	0.1
Salmonella sp.	91.4	400	> 8.04	< 0.48	7.6 minimum	0.1
Spores of Bacillus subtillis	90.6	400	3.25	1.30	2.0	0.3

Table EI: Summary of the inactivation results obtained on target organisms (Colas et al., 2008)

Regarding *Cryptosporidium* oocysts, reductions of approximately 5-log were obtained. For viruses (MS2 phages) reductions were lower and remained between 2- and 3-log for REDs of 480 J/m<sup>2</sup>.

The formation of nitrites was also studied. Figure E16 shows that traces of nitrites were measured in waters relatively low in nitrates (between 5 and 10 mg/L of  $NO_3$ )



Figure E16: Changes in nitrite production according to nitrate levels in the affluent (Colas et al., 2008)

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