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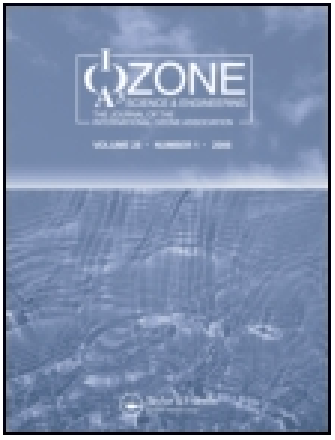

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The Basic Principles of UV-Disinfection of Water

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THE BASIC PRINCIPLES OF UV-DISINFECTION OF WATER

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Abstract

The disadvantage of chlorination of drinking water is the possible synthesis of toxic chlorinated fragments. In different cases UV can be an alternative to chlorination. The germicidal effectiveness of UV-radiation is in the 180-320 nm region with an optimum at 265 nm. Approximately 95% of the energy radiated by a low-pressure mercury arc is at the 253.7 nm line, so this source is the most effective one for germicidal applications. The germicidal effectiveness of a broadband source can be calculated. UV alone cannot decrease the concentrations of organic contaminants of the treated water. Quite promising are the systems where UV-radiation acts as a catalyst in oxidation reactions in order to decrease the organic contaminants.

Introduction

There is an increasing demand for quality water as drinking water, water for the food industry, breweries, soft drink manufacturers and as cooling water in sophisticated installations. Ultrapure water is required in the pharmaceutical industry and in high technology industries, such as the manufacture of semiconductors and integrated circuits.

For disinfection of drinking water, chlorination is widely used in the world until now. It is a very old and established method, since its introduction in the hospital by Semmelweis in Vienna, Austria in 1847. The advantages of chlorination are, for example, the long retention time and its killing efficiency for microorganisms at a low concentration level of about 0.3-0.5 mg/L in water.

Nowadays, there is increasing pollution of water sources by organic and inorganic compounds. During conventional chlorination of drinking water, dissolved organic molecules can be chlorinated. Many of these chlorinated fragments are toxic or suspected carcinogens, even in trace quantities (1-4). In particular for drinking water, the requirements for maximum contaminant levels for chlorinated compounds are in the ug/L range. This is one of the most important disadvantages of chlorination of drinking water at the moment. An old complaint about chlorinated water is its smell.

Because of the above-mentioned disadvantages of chlorination, a diminishing number of water sources without organic contaminants, and the increasing demand for quality water, the development of alternative water treatment technologies is of tremendous importance and a challenge to many scientists (5-8).

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UV, An Alternative To Chlorination

UV-radiation is optical radiation, and the same optical laws as used in the visible region can be applied to this part of the electromagnetic spectrum. For practical reasons, UV-radiation is divided into UV-A (400-315 nm, UV-B (315-280 nm), and UV-C (280-200 nm). The UV region below 200 nm is strongly absorbed by the air, and is called "vacuum UV". UV-radiation alone, in particular UV-C, is primarily used to destroy microorganisms.

The 253.7 nm line emitted by the low-pressure mercury discharge lamps (9-10) (Philips "TUV" lamps), can result in inactivation of microorganisms as protozoa, bacteria, molds, yeasts, viruses, fungi and algae.

The germicidal effect of UV in sunlight was found by Downes and Blount in 1877, and with the development of the mercury vapor lamp by Hewitt (1901), this UV technology was established. The germicidal effect of UV-radiation is based on photochemical reactions of biomolecules in the microorganism. These reactions can result in inhibited growth, or at higher doses in total inactivation.

Due to the high photon energy, many applications in the photochemical field use radiation sources in the ultraviolet region (11). When used in a proper way, UV-radiation can act as a strong catalyst in oxidation reactions to decrease the TOC (Total Organic Carbon) content in water treatment technology.

Figure 1 represents the photon energy as function of the wave-number or the wavelength in the UV- and visible region.

The disinfection of water by UV-radiation is dependent on the following parameters:

a. the emission spectrum of the UV-source;
b. the intensity of the irradiation and the exposure time;

c. the sensitivity of the micro-organism involved and the required survival ratio;
d. the performance of the reactor.

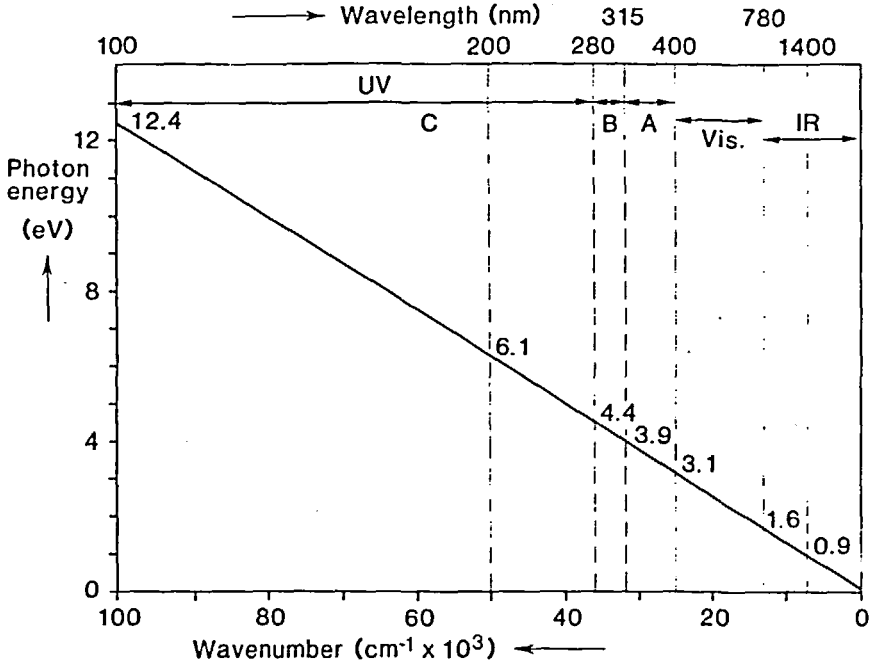

Figure 1. Photon energy as function of the wavelength.

Explanations of the above-listed parameters follow.

A. THE EMISSION SPECTRUM OF THE UV-SOURCE

The germicidal effectiveness is dependent on the wavelength of the UV-radiation, this action spectrum for germicidal activity is given in Figure 2. This relative effectiveness as a function of the wavelength has been determined for a common strain of E. Coli (13).

In this figure the effectiveness of the 253.7 nm mercury line (of the low-pressure "TUV" lamps) is shown, being 85% of the maximum at 265 nm.

It is seen that the maximum germicidal activity is at a wavelength of about 265 nm and decreases to longer and shorter wavelengths. The 253.7 nm line of a low-pressure mercury lamp (Figure 3) has an effectiveness of about 0.85, and with the conversion of 35% of the

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input power into the 253.7 line, this source is very effective for germicidal applications. This type of lamp is in fact used mostly in today's UV-treatment units for water disinfection. Also medium pressure mercury lamps (Figure 3), which are broad band sources, are suitable for germicidal purposes.

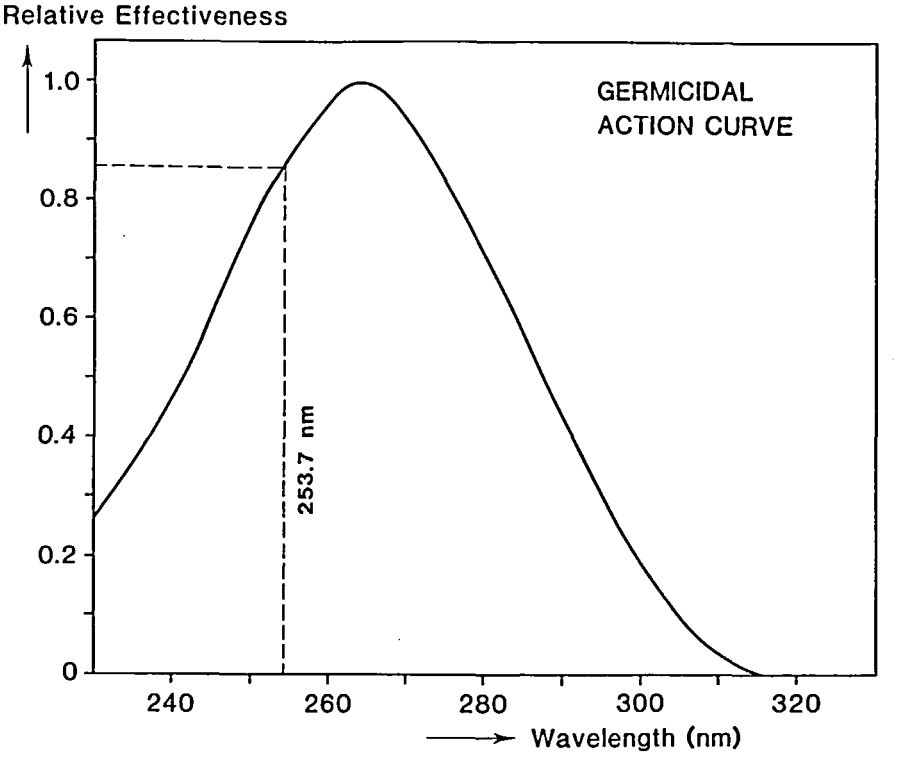

Figure 2. The germicidal action curve.

To determine the effectiveness of a broad band source, the irradiance (E_{eff}) at a distinct distance (depending upon the reactor design) has to be weighted against the action curve (Figure 2). The following weighting formula should be used:

$$\text{Effective Irradiance } (E_{eff}) = \sum_{210}^{315} E(\lambda) \times S(\lambda) \times \Delta\lambda \quad [W.m^{-2}]$$

where: $E(\lambda)$ is the spectral irradiance $W.m^{-2}$.
$S(\lambda)$ is the relative spectral germicidal effectiveness (see Table 1).
$\Delta\lambda$ integration of the spectral energy over a 5 nm region.

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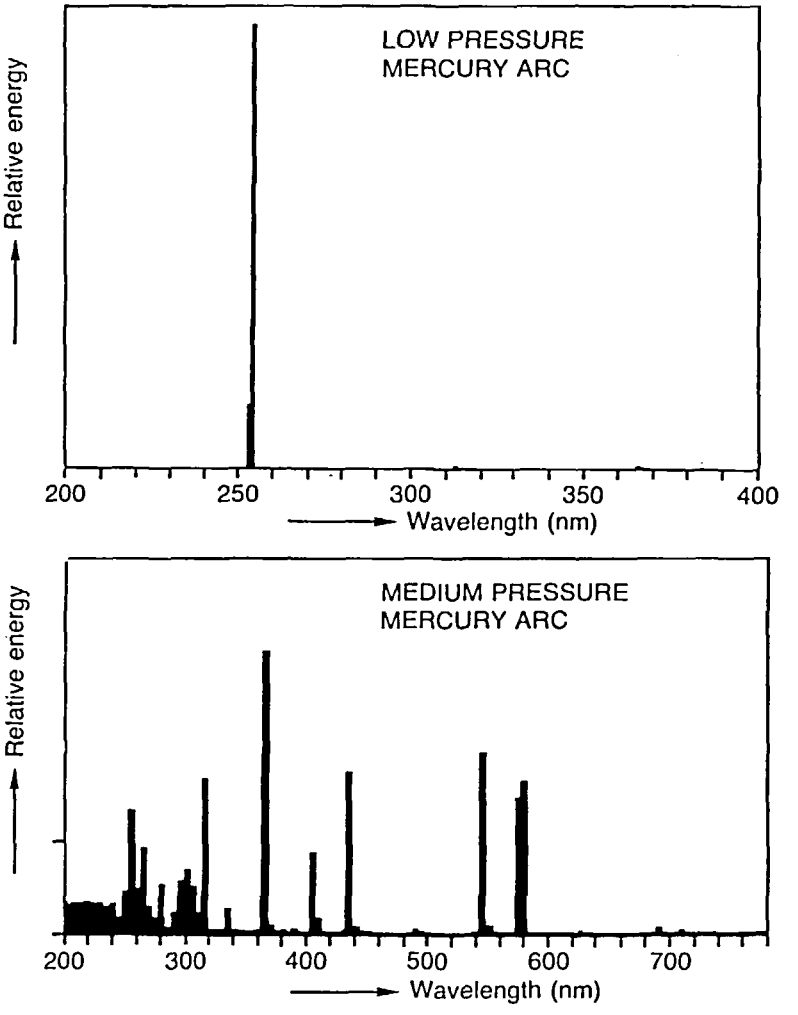

Figure 3. Emission Spectra.

For the calculation, the values of $S(\lambda)$ are normalized at 253.7 nm: $S_{253.7} = 1$. This is because the dose-values in Table II are determined at 253.7 nm.

For the case that the monochromator arrangement is adjusted to integrate 5 nm intervals, then in $\Delta\lambda$, the formula equals 1.

The effective irradiance, E_{eff}, multiplied by the time (in seconds) gives H_{eff}, the effective dose, in Joules.m^{-2}. Hence: $H_{eff} = E_{eff}$ x time gives the dose in J.m^{-2}.

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TABLE 1. S(λ) AS A FUNCTION OF THE WAVELENGTH. THE RELATIVE SPECTRAL EFFECTIVENESS FOR GERMICIDAL EFFECT

wavelength in nm	relative germicidal effectiveness
210	0.17
215	0.21
220	0.27
225	0.33
230	0.41
235	0.51
240	0.62
245	0.76
250	0.90
255	1.03
260	1.12
265	1.15
270	1.08
275	0.98
280	0.87
285	0.73
290	0.60
295	0.46
300	0.33
305	0.25
310	0.20
315	0.15

Taking a broad band source like a medium pressure mercury lamp (see Figure 3) and calculating the effective irradiance, this value is about half of the total unweighted irradiance in the 200-320 nm region.

B. INTENSITY AND EXPOSURE TIME

In order to produce a photochemical or a photophysical reaction, it is necessary that the object involved absorbs the radiation energy, according to Draper-Grotthus' Law (11). When the effect of the absorbed photon will change or break a chemical bond within the target, and a subsequent chemical reaction occurs, it is called a photochemical reaction. Many photochemical reactions have free radicals as intermediate products. These particles are very reactive and often produce chain reactions.

When the absorbed photon can only excite vibrational or rotational energy-levels, the only effect will be heat dissipation; this is called a photophysical reaction. This is particularly the case in the infrared region with low photon energies. A photophysical reaction in the UV-region occurs, when the excited state is losing its energy in the form of fluorescence, phosphorescence or radiationless heat dissipation.

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For radiation to have an effect, there must be an absorbing target. In the case of living cells, the DNA, RNA, and the proteins (20) are the targets with a strong absorption in the 260 nm region (see Figure 4). The total effect (the **response** of the system) is dependent on the total energy supplied (the **dose**).

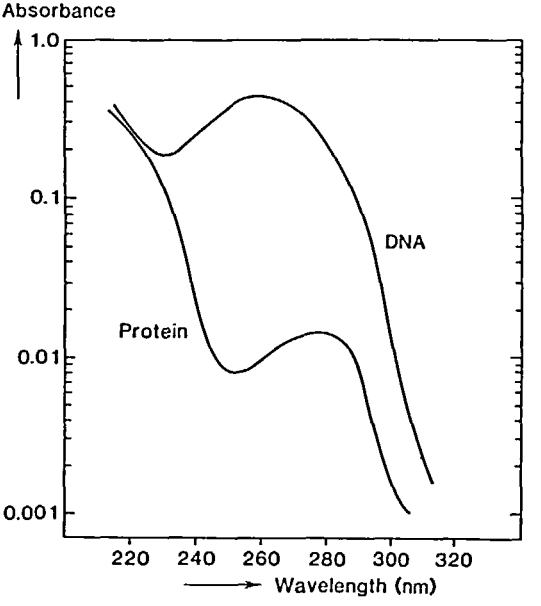

Figure 4. Absorbance of DNA and protein.

The **dose** (H_{eff}), being a quantity of energy, is, in the case of UV-radiation, the **irradiance** in [$W.m^{-2}$] at the target multiplied by the **time** in sec, to get the **dose** in [$J.m^{-2}$]. This **dose-response** relation for germicidal effect of UV normally is represented by survival ratio-curves (Figure 5).

When microorganisms are subjected to UV-radiation, they are not inactivated at once, but a constant fraction of the present living number dies in each increment of time (14-16, 20). The survival ratio is the fraction of the number of microorganisms initially present which survives at any given time. This survival ratio can be approximated by an exponential function of the product of exposure time and the effective irradiance intensity of the UV-radiation:

$$N/NO = \exp(-k.E_{eff}.t)$$

where:

N is the surviving number of microorganisms at time t

NO is the initially number of living species

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t is the time

E_{eff} is the effective irradiance of the UV-radiation

k is a constant depending on the sensitivity of the species

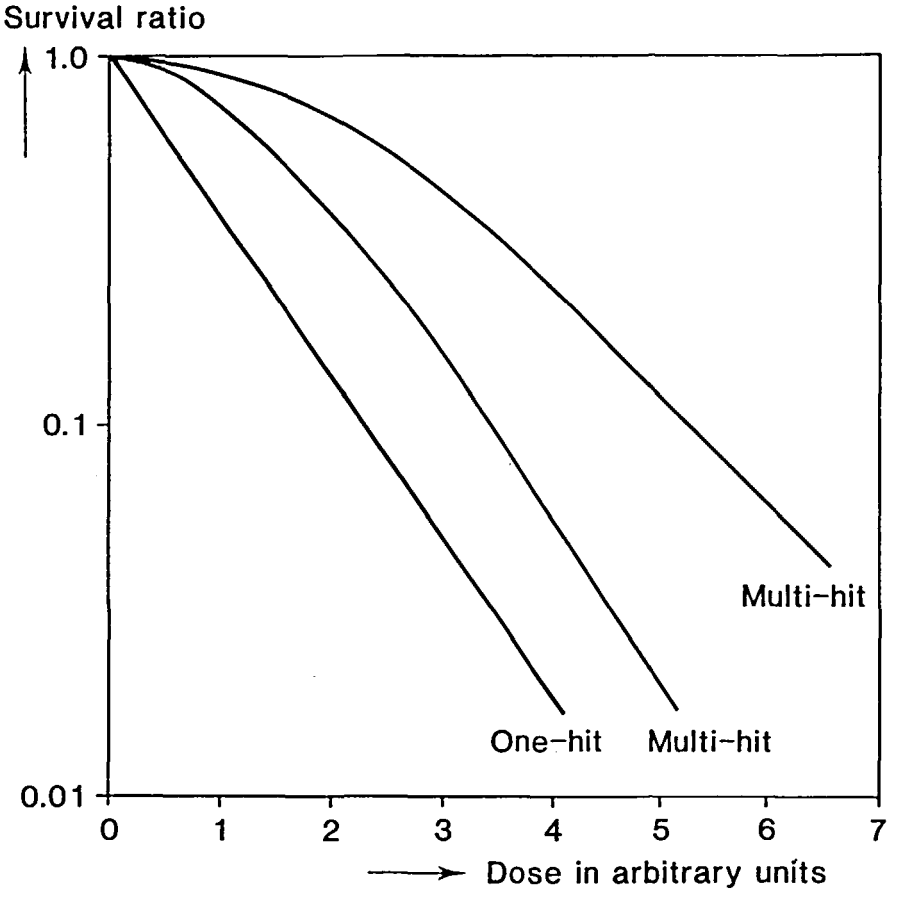

Figure 5. The survival curves.

The plot of N/NO vs the dose (E_{eff} x t) is a straight line on semilog paper. The practical consequence of this log-function is, that in order to decrease the survival ratio from 0.1 to 0.01, double the dose is needed. It is usual to tabulate the 0.1 survival ratio of the different species (see Table II).

If a survival ratio of 0.00001 is required, the tabulated dose has to be multiplied by 5.

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TABLE II. APPROXIMATE DOSE VALUES FOR A SURVIVAL RATIO OF 0.1 OF VARIOUS MICROORGANISMS AT 253.7 nm

Bacteria	Dose	Pseudomonas aeruginosa	55
Bacillus anthracis	45	Pseudomonas fluorescens	35
B. megatherium (veg.)	11	Salmonella enteritis	40
B. megatherium (spores)	27	S. typhosa-Typhoid fever	22
B. parathyphosus	32	S. paratyphi-Enteric fever	32
B. subtilis	70	S. typhimurium	80
(spores)	120	Sarcina lutea	197
		Serratia marcescens	24
Clostridium tetani	130	Shigella dysenteriae	
Corynebact. diphtheriae	34	Dysentery form	22
Eberthella typhosa	21	Shigella flexneri	
Escherichia coli	30	Dysentery form	17
Leptospira Spp.-Infectious		Shigella paradysenteriae	17
jaundice	32	Spirillum rubrum	44
Micrococcus candidus	61	Staphylococcus albus	18
Micrococcus piltonencis	81	Staphylococcus aureus	26
Micrococcus sphaeroides	100	Streptococcus hemolyticus	22
Mycobacterium tuberculosis	62	Streptococcus lactis	62
Neisseria catarrphalis	44	Streptococcus viridans	20
Phytomonas tumefaciens	44	Mycobacterium tuberculi	100
Proteus vulgaris	26	Vibrio comma-Cholera	34

Yeasts	Dose
Common yeast cake	60
Saccharomyces ellipsoideus (Bakers yeast)	60
Saccharomyces cerevisiae (Bakers yeast)	60
Torula sphaerica (as found in milk and cream)	23
Various algae	
Diatoms	3600-6000
Green algae	
Blue algae	
Protozoa	
Paramecium	640-1000
Worms	
Nematode eggs	400

Mould spores	Dose
Aspergillus amstelodami (meat)	667
Aspergillus flavus	600
Aspergillus glaucus	440
Aspergillus niger (bakeries)	1320
Cladosporium herbarum (cold stores)	600
Mucor mucedo (meat, fat, bread, cheese)	650
Mucor racemosus	170
Oospora lactis	50
Penicillium digitatum	440
Penicillium expansum	130
Penicillium chrysogenum (fruit)	500
Penicillium roqueforti	130
Rhizopus nigricans	1110
Scopulriopsis brevicaulis (cheese etc.)	800

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This straight line is an indication that one hit (one single quantum) is sufficient to inactivate the species involved. This type of curve is often observed in the surviving-dose relation of viruses (damage of single strand of RNA or DNA).

In general, when bacteria are exposed to UV, another type of curve is observed: the so-called "shouldered survival curve". This curve form is an indication that more than one hit is necessary for inactivation, but it can also be an indication that reactivation mechanisms are opposing the inactivation by UV. These reactivation mechanisms can be divided into "dark repair" and photoreactivation (17-20). These photoreactivation mechanisms are observed in experiments with bacteria in the 340-480 nm region.

In the medium-pressure lamp the emission is shifted towards the near UV- and visible region with the possible stimulation of photoreactivation mechanisms. It is theoretically possible to observe another survival curve, when a medium-pressure lamp is used instead of a low pressure tube, but in practice it can only be seen at very long exposure times.

The effect of the temperature in photoinactivation is negligible; an increase of 10°C gives an acceleration of only 1.05 to 1.1 times.

C. THE SENSITIVITY OF THE MICROORGANISM TO UV-RADIATION

The effective resistance of the many types of microorganisms to UV-radiation varies considerably from small doses for bacteria to very large doses to destroy algae. Moreover, the environment of the particular microorganism greatly influences the dose of radiation. In water, for instance, Escherichia coli being the most investigated bacteria from fecal origin, requires a dose being 5 to 10 times as great as for airborne E. coli to be destroyed for 90%.

The reason for a higher dose is that at high humidity in air, on moist surfaces and in water, the microorganism is protected by a thin water layer containing organic material which strongly absorbs the UV-C radiation.

Table II gives an impression of the sensitivity of a number of microorganisms to UV-radiation for the airborne situation.

In order to destroy a microorganism, UV-radiation has to hit one or more targets like DNA, RNA or enzymes in the inside of the microorganism. The required dose in the inside of the microorganism then is depending on the morphology of the microorganism involved. In general a gram-positive bacterium with a thick capsule of mureine is more difficult to destroy than a gram-negative bacterium with a thin capsula. Also, spore-forming bacteria are more difficult to destroy because of the very thick capsule of the spore. Algae need a hundred times more energy than bacteria. The sensitivity of many viruses is comparable with that of bacteria.

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Moreover a microorganism can be saved when it is in the shadow of a particle or other microorganisms during the exposure to UV-radiation.

It seems to be possible with ultrasonic techniques to separate the microorganisms from each other and from particles, resulting in a more efficient germicidal irradiation.

D. THE REACTOR PERFORMANCE

It is obvious that, when water has to be treated with UV, the average transmission of the water in the 200-300 nm region should be measured first. Depending on this parameter, the microorganisms to be treated, and the desired survival ratio, the effective UV dose can be calculated. Thus for water, the amount of radiation required to destroy a particular microorganism depends on the absorption coefficient (α) and the penetration depth (d), according to Lambert's law:

where:

$I_o = I\, e^{-\alpha d}$
I = intensity at depth d
α = absorption coefficient
d = penetration depth

The values of the absorption coefficient are between 0.007 and 0.01 cm^{-1} for distilled water, and between 0.02 and 0.1 cm^{-1} for mains water. Suspended solids can scatter the UV radiation and hence reduce the irradiance level. Dissolved organic matter and already low iron concentrations will increase the absorption coefficient.

In order to enable the lamps to operate at their optimum temperature, it is not allowed to immerse them in the water. It would be better that the lamp is first enclosed in a quartz container, which is separating the water to be disinfected from the lamp.

Installations may have the following forms:

a. One or more UV lamps enclosed in a quartz container which is surrounded by the water to be disinfected.

b. A quartz tube transporting the water, surrounded by a number of UV lamps in reflectors.

c. Irradiation by means of UV lamps installed in reflectors over the surface of the water flowing through a shallow tank.

In the design of the disinfection reactor, it is convenient to supply an overdose, taking into account the decreasing UV-output level of the lamps during their lifetime, and the correction factor for the wet situation. In the optical design, one should realize that every change of refraction index will give a partial reflection (a loss of intensity) of the UV-radiation according to Fressnel's Law.

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With the information of the UV-output supplied by the manufacturer of the lamps and a radiometer with a detector provided with a cosine-correction filter, a sufficient assessment of the intensity is possible.

Final Remarks

In the preceding discussion the inactivation of microorganisms is described. Depending on the clarity of water in the 200-300 nm region, UV-radiation can be very effective in destroying microorganisms.

A disadvantage of UV-treatment is that the material of the destroyed organisms and other contaminants will stay in the water.

In those applications with a short retention time (one of the disadvantages of UV) and an acceptable level of contaminants, disinfection with UV-radiation can be a sufficient water treatment technology. In other applications, where total mineralization of the organic contaminants is a requirement, oxidation processes must be used.

In the last decade, much effort has been applied to replace chlorination by another effective oxidative reagent. The demands made upon the reagent are that it is fading away or of a minor hazardous character after water treatment.

Quite promising are the systems in which UV radiation acts as a catalyst in oxidation reactions with ozone or hydrogen peroxide or both to decrease the TOC (21-29). With both reagents the active hydroxyl free radical can play an important role in the oxidation reaction rate.

In discussing the UV/ozone system, it is worthwhile to mention that the ozone to be used can be made by treating air by UV-radiation too.

Parallel with the effect of the temperature on reaction rates is the availability of another source of energy for "activating the chemical reaction". Usually this energy will be supplied in the form of thermal energy, as described by Arrhenius in defining the energy of activation. In photochemical reactions, this energy of activation is supplied in the form of photons. This primary reaction is independent of the temperature, and because of this feature, is a wonderful catalyst in these oxidation reactions.

Quantitative rate constants for photolysis of organic substances, in particular of high refractory contaminants, are still scarce. A lot of experimental work has to be conducted to evaluate reasonable kinetic models of these reactions, which are very specific for the species involved. A purely theoretical approach without experimental evidence is difficult due to the lack of knowledge about the intermediates and chain or non-chain mechanisms.

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In experimental work, the determination of the quantum yield, Q, for a given process is a good starting point:

$$Q = \frac{\text{number of reacting molecules}}{\text{number of absorbed photons}}$$

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Key Words

UV Radiation, Disinfection, Drinking Water Treatment, Fundamental Principles, Ozone/UV Radiation, Ozone/Hydrogen Peroxide

Résumé

Le désavantage de la chloration des eaux de consommation est la formation possible de composés chlorés toxiques. Dans differents cas, les radiations UV peuvent constituer une alternative à la chloration. L'efficacité germicide des radiations UV est dans la région 180-320 nm avec un optimum à 265 nm. Environ 95% de l'énergie irradiée par un arc à vapeur de mercure basse pression se trouve 253.7 nm. Par conséquent, c'est la source la plus efficace pour les applications germicides. L'efficacité germicide d'une source à large bande peut être calculée. Les UV radiations seuls ne peuvent pas dégrader les polluants organiques de l'eau traitée. Les systèmes dans lesquels le rayonnement UV agit comme un catalyseur dans les

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réactions d'oxydation pour la réduction des polluants organiques sont très prometteurs.

Zusammenfassung

Chlorung von Trinkwasser hat den Nachteil, dass Synthese von giftigen Chlorverbindungen möglich ist. In verschiedenen Fällen kann UV Strahlung eine Alternative zu Chlorbehandlung sein. Die keimtötende Wirkung von UV-Strahlung wird im Bereiche von 180-320 nm gefunden, mit einem Optimum bei 265 nm. Etwa 95% der von einem Tiefdruck Quecksilberentladungsbogen ausgestrahlten Energie befindet sich auf der 253.7 nm Linie, so dass diese Quelle für keimtötende Zwecke am meisten geeignet ist. Die keimtötende Wirkung einer Breitbandquelle lässt sich berechnen. Die organischen Verunreinigungen desbehandelten Wassers können mit UV allein nicht beseitigt werden. Aussichtsreich sind jene Systeme, in denen UV als Katalysator in Oxydationsreaktionen zur Verminderung des organischen Gehalts benutzt wird.

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