

## THE EFFECT OF MICROWAVE RADIATION ON WASTE TREATMENT IN A REACTOR WITH A BIOFILM

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**Abstract:** This paper presents the results of research into the effect of microwave radiation on waste treatment in a reactor with a biofilm. 2.45 GHz microwave radiation was supplied to the reactors placed inside a microwave chamber. The radiation was generated by magnetron and the amount of radiation was controlled by varying the times of alternating phases of radiation and treatment. The study was conducted in three arrangements of alternating phases: 7 s radiation and 10 min treatment; 7 s radiation and 5 min treatment; 25 s radiation and 10 min treatment. The results obtained in the study show that microwave radiation affects the process of biological waste treatment not only through heating but also through its athermal properties. An increase in the effectiveness of the treatment was particularly visible in the microwave action in nitrogen removal.

### INTRODUCTION

The rate of waste treatment processes which take place in a biofilm reactor depends on a number of parameters: the amount and activity of the biofilm, the size of the contact surface between the waste and biofilm and the mass exchange rate between the biofilm and waste. Additionally, the final purification effect is connected with the effectiveness of internal diffusion, external diffusion and biochemical transformations of organic compounds in cell metabolism.

The size of diffusion (the amount of transferred substance), is the result of concentration gradient, the surface and the factor of proportionality – also called the diffusion coefficient. The value of the coefficient is affected by physical conditions, with the temperature being the predominant factor. By affecting the diffusion, the temperature influences the kinetics of a number of unit processes of waste treatment, e.g. gas exchange, substrate solubility, liquid mixing.

The rate of biochemical decomposition of waste depends on the activity of microorganisms (precisely – on the enzymatic reaction rate). The ambient conditions, i.e. the substrate concentrations, temperature, redox potential, osmotic pressure and the presence of enzymes activating and inhibiting substances have an influence on the activity of enzymes.

The relationship between the reaction rate and temperature is expressed by the van 't Hoff-Arrhenius equation. A transformed form of the equation which compares the reaction rates at different temperatures is commonly used.

$$k_{T_1} = k_{T_2} e^{\left[ \left( \frac{E}{RT_1 T_2} \right) (T_1 - T_2) \right]} \quad (1)$$

where:

$k_{T_1}$  – reaction rate constant at the temperature  $T_1$  in variable units, depending on the reaction order,

$k_{T_2}$  – reaction rate constant at the temperature  $T_2$  in variable units, depending on the reaction order,

$E$  – activation energy [J·mol],

$R$  – universal gas constant [8.314 J·mol·K],

$T_{1,2}$  – temperature [K].

A transformed form of the equation is more commonly applied in modeling the processes of waste treatment:

$$k_{T_1} = k_{20} \theta^{(T_1 - T_{20})} \quad (2)$$

where:

$k_{20}$  – reaction rate constant at the temperature 293K (20°C) in variable units, depending on the reaction order,

$\theta$  – temperature coefficient,

$T_{20}$  – referenced temperature 293K (20°C).

The effect of temperature on the enzyme-catalyzed reaction rate is decided by two processes. An increase in temperature is accompanied by an enzyme-catalyzed reaction, which is consistent with the van 't Hoff-Arrhenius equation. However, because of the protein character of enzymes, the relationship is only valid below a certain temperature value. An increase in temperature above the optimum value for a given enzyme brings about its denaturation.

Assuming that optimizing the thermal conditions can improve the effectiveness of waste treatment processes, research was initiated into applying microwave radiation to this end. Microwave heating is selective. Its direct action causes an increase in the temperature of only those bodies whose dielectric properties allow them to absorb it [12]. These relationships are useful in trickling filters. Microwave radiation makes it possible to deliver energy directly to the biofilm (a radiation absorbing body). When the packing material for the reactor is made of plastic, which is transparent to radiation, the loss of radiation caused by radiation absorption is prevented. Applying a trickling filter makes it possible to heat up only biomass, with limited energy absorption by the treated waste.

## EXPERIMENTAL PROTOCOLS

The experiment was conducted with use of a dripping reactor with a biofilm, placed inside a chamber where it was exposed to microwave radiation. The casing and packing material for the reactor were made of material transparent to microwaves. The reactor's sensitive volume was  $V = 145 \text{ cm}^3$ , the packing specific surface  $s = 202 \text{ m}^2 \cdot \text{m}^{-3}$  and the theoretical active surface of the reactor was  $F = 0.029 \text{ m}^2$ .

The radiation was generated in a magnetron from where it was sent to the reactor chamber through a waveguide. The generator's total power was 800 W, its efficiency was 52% and it produced radiation at a frequency of 2.45 GHz. The magnetron emitted

radiation with a constant efficiency and the amount of energy supplied to the reactor was controlled by changing the times within the operation-break cycles. The operation of the microwave generator was synchronized by feeding the waste to the reactor. Before starting the generator, the feeding of waste to the reactor was cut off. Thanks to this, the microwave energy was absorbed primarily by the biofilm and not by the waste being treated. After the radiation phase ended, the waste dispensing pump restarted and the system returned to the purification phase (Tab. 1).

Table 1. The temperature inside the bioreactor in relation to the radiation input

Radiation phase/Treatment phase	The amount of microwave energy [W·s]	The temperature following the radiation phase [°C]	The temperature following the treatment phase [°C]
7 s/10 min	1.2	22.5	21.3
7 s/5 min	2.5	20.9	20.1
25 s/10 min	4.3	36.3	32.6

\* the amount of energy input stands for the total supplied power divided by the total work time of the one reactor.

The microwave radiation caused the temperature inside the biological reactor to rise. The temperature of the biofilm was the highest just after the work phase of the microwave generator ended and depended on the amount of supplied radiation. It then dropped as the purification phase continued (Tab. 1). The temperature of the control reactor and the environment was kept at a constant level of 20°C.

The study was conducted with use of municipal waste. It was taken directly from the municipal collector at the same hour once daily. After being collected, the waste was left for 0.5 hour for sedimentation and chemical analyses were conducted (Tab. 2).

Table 2. The values of pollution indexes in raw sewage

Index	Unit	Mean value	Standard deviation
COD	[mg O <sub>2</sub> ·dm <sup>3</sup> ]	220	21.1
BOD <sub>5</sub>	[mg O <sub>2</sub> ·dm <sup>3</sup> ]	175	14.4
Total nitrogen	[mg N <sub>org</sub> ·dm <sup>3</sup> ]	45	5.6
Ammonium nitrogen	[mg N-NH <sub>4</sub> ·dm <sup>3</sup> ]	25	5.3
Total phosphorus	[mg P <sub>org</sub> ·dm <sup>3</sup> ]	18	3.8
Phosphates	[mg P-PO <sub>4</sub> ·dm <sup>3</sup> ]	12	3.7
Total suspended matter	[mg·dm <sup>3</sup> ]	240	81.5

The experiment was conducted periodically. Every 24 hours, the entire volume of waste in the impounding reservoir was replaced. The waste from the impounding reservoir was fed to the biological reactor and then returned to it (Fig. 1). The hydraulic load  $q = 0.15 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$  was applied, as a result of which the whole amount of waste in the reactor flowed through the biological reactor 21 times in 24 hours. The organic pollution load on the surface of the reactor with the pollution, expressed as COD, was close to  $A' - 8.6 \text{ g O}_2 \cdot \text{m}^2 \cdot \text{d}^{-1}$ . The experiment was conducted for 30 days for each series which differed by the amount of radiation supplied to the reactors (Tab. 1).

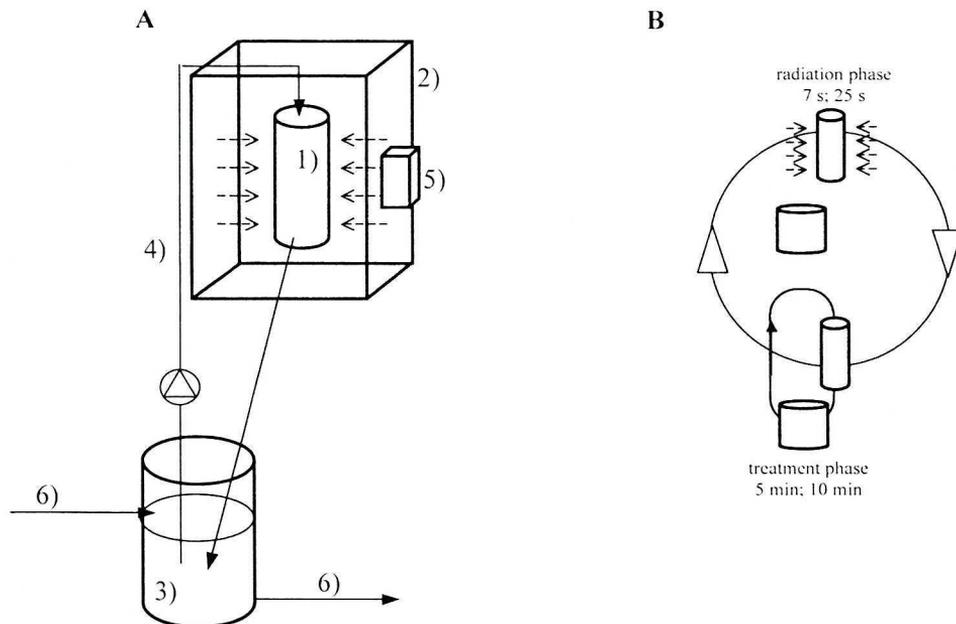


Fig. 1. A – diagram of the experiment layout: 1) biological reactor, 2) microwave chamber, 3) retention reservoir, 4) sewage supply to the reactor, 5) microwave generator, 6) sewage replacement in the retention reservoir; B – diagram of the operation of the experiment arrangement

In the purified waste taken each day, the organic substance, expressed as COD, was determined as well as the nitrogen compound concentration: nitrogen in ammonium  $N-NH_4$ , nitrogen in nitrates  $N-NO_3$ , nitrogen in nitrites  $N-NO_2$ , Kjeldahl's nitrogen (ammonium nitrogen + organic nitrogen). Additionally, the biomass yield was calculated. The quantity of biomass was analyzed inside the reactor at the beginning and at the end of the measuring period, including the amount of excess biofilm in the effluent. Based on the analyses, the effect of the applied microwave radiation on the effectiveness of organic compound and nitrogen compound removal was measured.

The results were analyzed statistically with use of a single-factor analysis of variance, at the assumed level of significance of  $p < 0.05$ . The normality of distribution was confirmed by the W Szapiro-Wilk test, while the hypothesis of the variance uniformity was verified with Leveney's test. The differences between the average values of various groups were examined with Tukey's HSD test. The reaction rate constants were determined by the method of regression and based on the experimental data.

As in the control, tests were conducted with a reactor which was not exposed to microwave radiation and which worked in identical hydraulic conditions, with the same load of pollution. Inside the control reactor, the temperature was  $20^\circ C$  and hot air was used for heating.

## RESULTS

The experiment provided data for determining the effect of the applied microwave radiation on the effectiveness of waste treatment. This paper presents the basic parameters which determine the efficiency of biological waste treatment. The efficiency of the process can be described by the efficiency of organic compound removal, nitrogen compound removal and biomass increase. Nitrogen compounds were removed as a result of a biochemical reaction cascade. Normally, three consecutive phases are mentioned: ammonification, nitrification and denitrification. Nitrogen transformations were described by determination of nitrification efficiency and nitrogen loss when the denitrification process and biomass increase were taken into account. The effectiveness of organic compound removal was expressed as a decrease of COD in the waste being treated. The contribution of oxidation and biomass synthesis to the process of organic substance removal was also determined.

Microwave radiation had a considerable effect on the efficiency of organic substance removal from waste only in one of the three doses used in the experiment. When 2.4 W·s was supplied to the reactor in the system in which the reactor worked for 7 seconds every 5 minutes, the reactor's efficiency increased by 7.3% as compared to that of the control reactor. When the amount of radiation was smaller – 1.2 W·s – a low efficiency increase of 2.3% was observed. However, when the radiation was supplied to the reactor for 25 seconds every 10 minutes (4.3 W·s), this resulted in a slight drop in the efficiency of organic substance removal by 3.5% (Fig. 2). Statistically significant differences were not found among the series, using a level of significance of  $p < 0.05$ . Although the effect of microwaves on the final efficiency of organic substance removal was slight, it was significant in the unit processes. Microwaves caused an increase in the contribution of oxidation in organic substance removal, while the significance of biomass synthesis decreased. As a result of supplying as little as 1.2 W·s of radiation to the system, more than 10% more organic substance was removed due to oxidation. When twice as much energy (2.4 W·s) was supplied, oxidation was responsible for removing 78.4% of organic matter, while in the control reactor the value was merely 54% (Fig. 2).

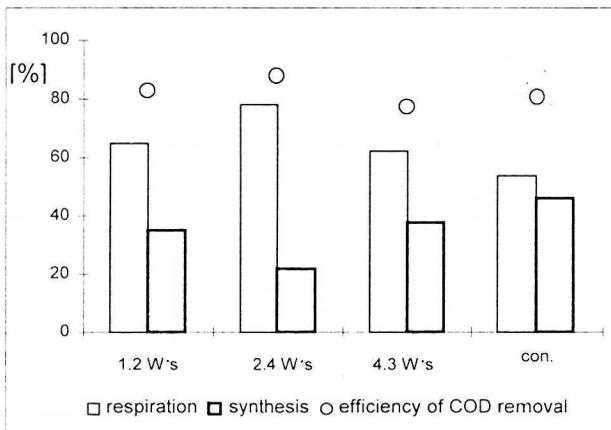


Fig. 2. Dependence of efficiency of COD removal and participation respiration and synthesis in this process on microwave amount

The most significant effect was observed when microwave radiation affected the efficiency of the nitrification process. As a result of supplying a mere 1.2 W·s of microwave energy, the efficiency of nitrification increased by 32% as compared to the control reactor. A larger amount of energy (2.4 W·s) caused a similar effect – an increase by 30.4%. There were no statistically significant differences between the series. Supplying radiation for 25 seconds every 10 minutes caused the efficiency to drop by 19.5% in relation to the control reactor, although the amount of supplied microwaves was the largest (4.3 W·s) (Fig 3). Nitrification efficiency in this series was statistically lower than in the other series.

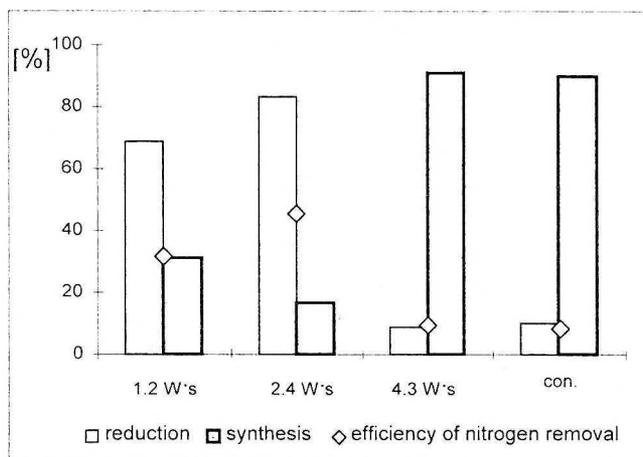


Fig. 3. Dependence of efficiency of nitrogen removal and participation reduction and synthesis in this process on microwave amount

An analysis of the effect of microwaves on the efficiency of nitrogen removal revealed a 23% increase compared to the control reactor when 1.2 W·s of microwave energy was supplied to it. However, an increase in the amount of radiation supplied to the reactor to 2.4 W·s caused the removal of 45.4% of the total nitrogen from the raw waste, while in the control reactor it did not exceed 8.6%. These differences were statistically significant at the assumed level of significance of  $p < 0.05$  with the use of Tukey's test. In the third arrangement of power and time of radiation (4.3 W·s), the nitrogen removal was similar to that in the control reactor. It is even more interesting to analyze the effect of microwaves on the contribution of various processes to nitrogen removal. Reduction of nitrates to gaseous forms dominated in those series where 1.2 W·s and 2.4 W·s of radiation was supplied to the system, accounting for as much as 83.1% of the transformations, while in the other cases almost all the nitrogen was removed by incorporating it in the newly created biomass (Fig. 4). In the control reactor, 90.1% of the removed nitrogen was incorporated in the new biomass, with 9.9% denitrified.

Among the key factors in the analysis of the process of biological waste treatment there are the thermal conditions in the reactor during the consecutive parts of the experiment. The study was conducted with three various amounts and time arrangements of radiation supply to the system. In the series when the radiation was supplied for 7 seconds every 10 minutes, the temperature rose to 21.2°C and then dropped to 20.0°C. The

average temperature was about  $20.5^{\circ}\text{C}$ , which was higher by  $0.5^{\circ}\text{C}$  than in the control reactor. In another series, when the generator worked for 7 seconds every 5 minutes, the maximum temperature in the reactor was  $22.5^{\circ}\text{C}$ . At the end of the purification phase, after 5 minutes, it dropped to  $21.7^{\circ}\text{C}$ , the average value being  $22.0^{\circ}\text{C}$ . The highest temperature differences were observed when the radiation was supplied for 25 seconds every 10 minutes. At the end of the radiation phase, the temperature inside the reactor rose to  $36.3^{\circ}\text{C}$  and after the 10-minute purification phase it dropped to  $32.6^{\circ}\text{C}$ , with the average value equal to  $33.7^{\circ}\text{C}$  (Fig. 5).

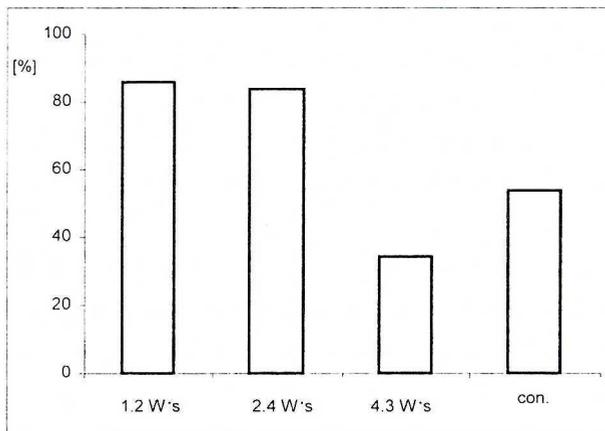


Fig. 4. Dependence of nitrification efficiency on microwave amount

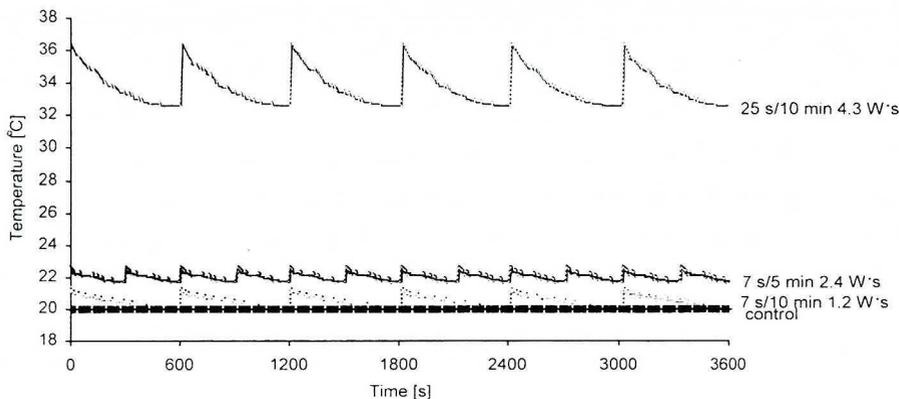


Fig. 5. Changes of temperature inside reactors during experiment

## DISCUSSION

The effect of microwave radiation on the efficiency of waste treatment, observed in the experiment, depended on the amount of supplied radiation and on the time arrangement in which it was supplied. The efficiency of pollution removal changed very little as a

result of radiation. The maximum increase observed in the efficiency of the process was by about 7.3% in the series when 2.4 W·s of microwave energy was supplied, and the average temperature inside the reactor was higher by 2°C than inside the control reactor. Such results were consistent with the expectations. The literature contains numerous comments on the relatively small effect of change in the temperature on the activity of heterotrophic bacteria in the mesophilic temperature range. Kadlec and Reddy [5] point out that the reactions of microorganisms depend on temperature to a greater extent at its lowest values (below 15°C) than in the optimum range between 20 and 35°C, and a change in temperature in this range does not have a significant effect on BOD<sub>5</sub> removal. A study into the colony forming abilities of heterotrophic bacteria conducted by Mayo and Noike [6] showed that the process rate is the same at 20°C and at 35°C. The authors emphasized that under stable conditions, heterotrophic bacteria did not react to temperature changes within the range from 10 to 20°C. The number of heterotrophic bacteria at 10 and 20°C was similar. Pabello *et al.* [7] found that a 21% increase in carbon compound removal took place between 10 and 20°C, while a further increase to 30°C brought about only an increase by 13%. In the current study, a temperature increase to the mean value of 33.7°C by radiating the waste for 25 seconds every 10 minutes reduced the amount of removed organic matter. This effect can probably be attributed to large temperature differences. During the 25 second radiation phase, the temperature inside the reactor rose by 3.7°C, to drop subsequently during the purification phase. According to numerous authors, an adverse effect of unstable temperature on the efficiency of the removal of organic matter expressed as COD is observed mainly in the processes which take place at higher temperatures. Tripathi and Allen [13] found that in order to avoid the adverse effect of temperature shock, an adaptation period of at least one week must be applied when the temperature is increased by 5°C.

Microwave radiation of the biofilm significantly affected nitrogen removal from the waste. Thanks to the action of 1.2 W·s and 2.4 W·s of radiation, the nitrogen was removed, not only by biomass synthesis, but primarily as a result of the reduction of oxidized forms. Denitrification was possible mainly thanks to creating anoxic areas deep inside the biofilm, which was primarily due to microwave heating. Microwave energy causes a body to heat up in its whole volume. Considering the low thickness of a biofilm in relation to the depth of radiation penetration, it can be supposed that the biofilm was heated up uniformly. The relationship between creating the anoxic zones and the temperature of the biofilm was corroborated by a study conducted by Hao *et al.* [3], who found the oxygen penetration depth increased with a decrease in temperature. However, it must be noted that in the part of the experiment in which 4.3 W·s of radiation was supplied to the system, only a limited denitrification was observed, despite favorable temperature conditions and the possibility of creating anoxic zones. In this case, the nitrogen removal may have been limited by a low efficiency of nitrification. The possibility of simultaneous nitrification and denitrification in a settled biomass has been widely discussed in literature [2, 4, 11, 14].

The effect of microwave radiation on the process of waste treatment in the biofilm reactor can be explained by the presence of a thermal effect. A small increase in the degree of organic compound removal can be explained by the relatively low sensitivity of heterotrophic bacteria to temperature changes, while an increase in nitrogen was caused by the fact that temperature changes affected the creation of anoxic zones. However,

numerous phenomena were observed during the study which cannot be explained by the temperature effects which accompanied the radiation. The lowest of the applied amounts of radiation (1.2 W·s) caused the efficiency of nitrification to increase by as much as 32%, with the average temperature inside the reactor higher than that inside the control system by only 0.5°C. Surprisingly, such a small temperature increase was accompanied by a considerable increase in the amount of oxidized organic compounds and in the efficiency of nitrification. A temperature rise may have influenced the process of nitrification, but the efficiency of nitrogen removal in the control reactor, with a temperature lower by mere 0.5°C, was equal to one third of the former and denitrification was hardly observable. These unexpected effects were probably caused by the athermal properties of microwaves. Electromagnetic radiation can stimulate enzyme activity. Banik *et al.* [1] provide numerous examples to show the possibility of an athermal positive effect of microwaves on microorganisms. Parker *et al.* [10] examined the effect of microwaves on hydrated lipase. It was found that when the reaction environment was heated with microwaves, the enzymatic reaction rate increased 2- or 3-fold in relation to conventional heating. In discussing the effect of microwave radiation on biological matter, Ponne and Bartels [9] referred to a number of studies in which both positive and negative effects on microorganisms were found. On the other hand, some authors have questioned the very existence of non-thermal microwave effects [10].

## CONCLUSIONS

- The effect of microwave radiation on the efficiency of organic matter removal was low and not statistically significant.
- A significant (over 30%) increase in the nitrification effectiveness with a slight temperature increase could suggest the athermal properties of microwave radiation.
- A large increase in denitrification efficiency in a radiation reactor with only a slightly higher temperature could also demonstrate the positive athermal effect of microwaves.

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## REFERENCES

- [1] Banik S., S. Bandyopadhyay, S. Ganguly: *Bioeffects of microwaves. a brief review*, *Biores. Technol.*, **87**, 155–162 (2003).
- [2] Chui P.C., Y. Terashima, J.H. Tay, H. Ozaki: *Performance of a partly aerated biofilter in the removal of nitrogen*, *Wat. Sci. Tech.*, **34**, 187–194 (1996).
- [3] Hao X., J.J. Heijnen, M.C.M. van Loosdrecht: *Model-based evaluation of temperature and inflow variations on a partial nitrification-anammox biofilm process*, *Wat. Res.*, **36**, 4839–4849 (2002).
- [4] Helmer C., S. Kunst: *Simultaneous nitrification/denitrification in aerobic biofilm system*, *Wat. Sci. Tech.*, **37**, 183–187 (1998).
- [5] Kadlec, R.H., K.R. Reddy: *Temperature effects in treatment wetlands*, *Wat. Environ. Res.*, **73**, 543 (2001).
- [6] Mayo A.W., T. Noike: *Effects of temperature and pH on the growth of heterotrophic bacteria in waste stabilization ponds*, *Wat. Res.*, **30**, 447–455 (1996).
- [7] Pabello L.V., A. Alardo-Lubel, C. Durana-De-Bazua: *Temperature effects on ciliates diversity and abundance in rotating biological reactor*, *Biores. Tech.*, **39**, 55–60 (1992).

- [8] Parker M.C., T. Besson, S. Lamare, M.D. Legoy: *Microwave radiation can increase the rate of enzyme catalysed reaction in organic media*, *Tetrahedron Letters*, **37**, 8383–8386 (1996).
- [9] Ponne C.T., P. Bartels: *Interaction of electromagnetic energy with biological material – relation to food processing*, *Radiat. Phys. Chem.*, **45**, 591–607 (1995).
- [10] Porcelli M., G. Cacciapuoti, S. Fusco, R. Massa, G. D'ambrosio, C. Bertoldo, V. de Rosa Zappia: *Non-thermal effects of microwaves on proteins: thermophilic enzymes as model system*, *FEBS Letters*, **402**, 102–106 (1997).
- [11] Siegirst H., S. Reithaar, G. Koch, P. Lais: *Nitrogen loss in a nitrifying rotating contactor treating ammonium-rich wastewater without organic carbon*, *Wat. Sci. Tech.*, **38**, 241–248 (1998).
- [12] Thostenson E.T., T.-W. Chou: *Microwave processing: fundamentals and applications*, *Composites, A*, **30**, 1055–1071 (1999).
- [13] Tripathi C.S., D.G. Allen: *Comparison of mesophilic and thermophilic aerobic biological treatment in sequencing batch reactors treating bleached kraft pulp mill effluent*, *Wat. Res.*, **33**, 836–846 (1999).
- [14] Watanabe Y., S., Masuda, M. Ishiguro: *Simultaneous nitrification and denitrification in micro-aerobic biofilms*, *Wat. Sci. Tech.*, **26**, 511–522 (1992).

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#### WPLYW PROMIENIOWANIA MIKROFALOWEGO NA OCZYSZCZANIE ŚCIEKÓW W REAKTORZE Z BŁONĄ BIOLOGICZNĄ

W pracy przedstawiono wyniki badań nad wpływem promieniowania mikrofalowego na proces oczyszczania ścieków w reaktorze z błoną biologiczną. Do reaktorów umieszczonych we wnętrzu komory mikrofalowej dostarczano promieniowanie mikrofalowe o częstotliwości 2,45 GHz. Jako źródło mikrofal wykorzystano pracujący ze stałą moc magnetron, natomiast sterowania ilością dostarczanego promieniowania odbywało się poprzez zmienne czasy trwania następujących kolejno faz napromieniowania i oczyszczania. Badania przeprowadzono w trzech układach następujących cyklicznie faz: 7 s napromieniowanie 10 min oczyszczanie, 7 s napromieniowanie 5 min oczyszczanie oraz 25 s napromieniowanie 10 min. oczyszczanie. W ten sposób dostarczano do reaktora promieniowania, odpowiednio w ilości 1,2 W·s, 2,5 W·s oraz 4,3 W·s. Uzyskane wyniki badań wskazują, że promieniowanie mikrofalowe wpływa na proces biologicznego oczyszczania ścieków nie tylko w wyniku klasycznego ogrzewania, ale i poprzez swoje właściwości atermiczne. Szczególnie w przypadku przemian prowadzących do usunięcia azotu obserwowane było istotne zwiększenie ich sprawności w wyniku oddziaływania mikrofal.