DIELECTRIC BEHAVIOR OF SLUDGE FROM WASTEWATER TREATMENT

CSABA BARTHAa, ALINA CARAMITUb, MONICA JIPAa*, DANIELA MARIA IGNATa, ATTILA TÓKOSa

ABSTRACT. In the present study, dielectric spectroscopy was used to assess the influence of ELF (1-200 Hz) electromagnetic fields over protein relaxation from activated sludge (approx. 99% water content), sampled from the biological tank of a domestic wastewater treatment plant. Dielectric loss (tgδ) was determined for both activated sludge samples and autoclaved samples. Experimental data analysis shows that biological samples (activated sludge) have different dielectric behavior than the sterile samples (sterilized sludge). Unlike sterile samples in which the function tgδ vs. frequency is continuous, in the activated sludge samples, this function presents two discontinuities specific to electrical resonances around the frequencies of 26.5 Hz and 50.1 Hz. This behavior indicates that under the influence of the measuring signal of 26.5 Hz and 50.1 Hz, biochemical processes are stimulated in activated sludge. Also, the significant changes in the number of charge carriers suggest that, at these frequencies, changes occur in the mechanism and kinetics of biochemical processes.

Keywords: wastewater, activated sludge, dielectric spectroscopy, dielectric loss, electromagnetic fields, ELF

INTRODUCTION

From the perspective of sustainable development, streamlining the wastewater treatment process is a priority issue [1-3], theoretically complex, and with particular technical and economic implications.

*Corresponding author: monica.jipa@icpebn.ro
The theoretical complexity of the problem is mainly due to the unique diversity of organic [4-5] and inorganic [7-9] (sometimes radioactive [10]) pollutants contained in industrial and domestic wastewater.

Commonly, most wastewater treatment plants are provided with a biological treatment stage, where pollutant removal is carried out through biologically active sludge with high biodiversity [11-16].

Due to their specificity, the biochemical processes are relatively slow, representing the rate-determining step of the treatment processes. For this reason, the share of energy costs related to the microbiological stage is usually over 60% of the total treatment costs [11]. Given these considerations, research and developments upon streamlining the biological stages of water treatment are currently of great importance.

Attempts to accelerate the microbiological purification step by using bioactivators are reported [17]. Lingvay et al. [9], shows the ability of some plant extracts to activate the metabolism of microorganisms (bacteria and molds) even in environments with xenobiotic pollutants, usually present in domestic wastewater.

On the other hand, several studies show that extremely low-frequency electromagnetic fields (ELF-EMF) influence the mechanism and kinetics of natural electrochemical processes [18], effects of accelerating corrosion processes, and microbiological degradation of various materials used in construction and installations - both metallic and oxide [19-22] as well as polymeric ones [23-26], have been reported. Also, ELF influences on the living matter have been reported, such as stimulation of some biotechnological processes [27, 28], changes in circadian rhythm [29-31], etc.

It has been found experimentally, that intense electromagnetic fields, higher than 15 V rms, lead to degradation by permeabilization of the cell membrane followed by leakage of the intracellular compounds [32, 33]. Also, in the study of the vacuolar H⁺-ATPase (V-ATPase) rotary enzyme biocatalytic activity behavior in ELF, maximum changes were found around the frequency of 86 Hz, explainable by synchronisms in the ion-pumping steps in individual enzymes via a hold-and-release mechanism [34]. Recent studies have shown significant changes in the biocatalytic activity, selectivity, and growth of yeast cultures [35-38]. Through dielectric spectroscopy technique and specific biochemical techniques on Aspergillus Niger behavior, it was found that mold responds selectively only at certain discrete frequencies in the ELF field [39]-depending on the applied ELF frequency, the metabolites change [42], and upon exposure in the range of 10-15 V rms of 50 Hz, the development (maturation) and multiplication rate (spore production) increase substantially [33].

Thus, in the current research, we aimed to assess activated sludge behavior in ELF (1-200 Hz) through the dielectric spectroscopy technique.
RESULTS AND DISCUSSION

According to [43], the results obtained, regarding the content of sedimentable suspended solids (activated sludge) in the investigated water sample are summarized in Table 1.

| Method     | Volumetric [cm³/dm³] | Gravimetric [g/dm³] |
|------------|----------------------|---------------------|
| Sample     | 1        | 2        | 3        | 4        | 5        | 1         | 2         | 3         | 4         | 5         |
|            | 235      | 250      | 240      | 260      | 255      | 4.8562    | 5.6008    | 5.5058    | 6.4012    | 6.0502    |
| Average    | 248      |           |           |           |           | 5.6828    |           |           |           |           |

Results from Table 1 show that the average content of sedimentable suspended solids of the investigated water sample is 248 cm³/dm³, respectively 5.6828 g/dm³ after drying at 105 °C (approx. 99.4% water content).

The results obtained through the dielectric spectroscopy technique performed on the wet sediment of activated sludge, respectively, the evolution of dielectric loss ($\varepsilon''$) depending on the applied frequency (in the range 1-200 Hz) are presented in Figure 1. Figure 1 shows that the evolution of dielectric loss depending on the applied frequency presents discontinuities at 26.5 Hz and 50.1 Hz.

The dielectric loss $\varepsilon''$ is the ratio between the real component $\varepsilon'$ and the imaginary one $\varepsilon''$ of the dielectric permittivity (1):

$$\varepsilon'' = \frac{\varepsilon''}{\varepsilon'}$$

(1)

The imaginary component of the dielectric permittivity is determined by the electrical conductivity of the medium $\sigma$ at a given frequency $f$, respectively (2):

$$\varepsilon'' = \frac{\sigma}{2\pi f}$$

(2)

Under these conditions, the discontinuities in Figure 1 indicates that under the influence of the electric field generated by the measuring signal, dynamic processes (1) occur near frequencies of 26.5 Hz and 50.1 Hz- such as protein condensation/hydrolysis- which suddenly changes the number of charge carriers (and implicitly of $\sigma$) in the investigated sample (biomass-activated sludge). Similar results have been reported on Aspergillus Niger cultures [39].
It is noted that α amino acids with aliphatic groups $R_1$ and $R_2$ from (1) - depending on the size of these groups, have relaxation times and, consequently, different resonance frequencies. Under these conditions, following the application of an ELF field, depending on the frequency of the applied field, certain α amino acids resonate (depending on the volume and shape of the molecule, the dipole moment, the viscosity of the intracellular medium, etc.- there is practically a selective "agitation" at a given frequency of the polar molecule). Additionally, the observed dynamic processes near these frequencies may be due to the changes in charge distribution upon the interaction between the EMF fields with the cells' molecular system. Proteins present a specific amino acid sequence, formed by linking the α-carboxyl group of one amino acid to the α-amino group of another amino acid with a peptide bond (Scheme 1).

Because of the different charge distribution of the constituting polar and charged molecules, proteins are polarizable materials with specific electrostatic properties, contributing to the proteins electric and dielectric properties.
When an electrical field is applied, the randomly orientated polar molecules, including positive and negative charged molecules within the protein, rotate, move, align and polarize, triggering structural changes in proteins [40]. This displacement of charge distribution that drives such conformational changes plays an important role in membrane transport proteins and could explain the DNA's stimulation that initiates protein synthesis. Following the study of the electric and magnetic field influence on the Na⁺/K⁺ ATPase enzyme activity, results suggest that upon application of an electrical field, changes occur in the distribution of electrical charges, similar to the ion-binding activation mechanism. Studies also reveal that electric and magnetic fields act at different sites on the ATPase, changing the surface charge density and affecting ion transport across the membrane [41]. Thus, if certain frequencies stimulate the charge transfer process, previously reported results could be explained [23-26, 33, 35-38].

For a better highlighting of the electrical oscillations produced by the measurement signal applied to the investigated biomass (Figure 2), the evolution of derivative $\Delta \frac{tg\delta}{\Delta f}$ depending on frequency is presented.

**Figure 2.** The evolution of $\Delta tg\delta/\Delta f$ as a function of the active sludge frequency

Figure 2 suggests that around frequencies of 26.5 Hz and 50.1 Hz electrical resonance phenomena occur, explainable by sudden changes in the concentration of charge carriers in the investigated samples.

In Figure 3, the evolution of dielectric loss ($tg\delta$) depending on the applied frequency in the range 1-200 Hz, obtained on the sterilized sludge sample by autoclaving at 105 °C for 30 minutes, is presented.

Figure 3 shows that the evolution of the dielectric losses depending on the applied frequency on the sterilized sludge is a continuous function without discontinuities/electrical resonances, as presented in Figure 1 and 2. This observation suggests that in the sterilized material (denatured proteins), under the influence of the measuring signal, there are no processes by which the number of carriers suddenly changes.
By comparing Figure 1 with Figure 3, it was found that the $\tan \delta$ values of the sterilized material are systematically higher (up to 70 times) than of the activated sludge. This finding can be explained by the fact that after heat-based sterilization, cytoplasmic proteins are denatured, disturbing the natural balance (1) (hydrolysis no longer occurs), and the cell membranes disintegrate, discontinuing the activity of the ion pumps, which leads to the release of ionic species (charge carriers) from the cytoplasm and thus to the corresponding increase in conductivity $\sigma$.

These comparative results (activated sludge/sterilized sludge) suggest that dielectric relaxation is a specific feature of living matter, where the dynamic processes of hydrolysis/condensation of proteins, are not degenerated.

![Figure 3. Dielectric losses on sterilized sludge suspension](image)

Therefore, according to [33] and [39], it is considered that by applying an electric field of up to 15 Vrms/cm around the frequency of 26.5 Hz or 50.1 Hz, significant changes may occur in the activity/metabolism of the microorganisms from the activated sludge. These findings suggest that by applying an electric field in the ELF range, biological water purification processes can be controlled, both quantitatively (accelerating the process) and qualitatively (the share of metabolites formed - biogas [13], biohydrogen [12], etc.).

CONCLUSIONS

Through dielectric spectroscopy technique in the range 1-200 Hz, dielectric loss $\tan \delta$ were determined for both activated sludge sediment sampled from a wastewater treatment plant and autoclaved sludge sediment.
After comparing the experimental results, it was found that living matter samples (activated sludge) have different dielectric behavior than the sterile samples (sterilized sludge), respectively:
- in sterile samples, the function $\tan \delta$ vs. frequency is continuous, without discontinuities;
- in activated sludge samples, the function $\tan \delta$ vs. frequency has two specific discontinuities specific to electrical resonances around the frequencies of 26.5 Hz and 50.1 Hz, respectively.

This behavior indicates that under the influence of the measurement signal of 26.5 Hz and 50.1 Hz, respectively, in activated sludge, biochemical processes are stimulated that take place through significant changes in the number of charge carriers - which suggests that at these frequencies, there are changes in the mechanism and kinetics of biochemical processes from the activated sludge.

EXPERIMENTAL SECTION

The investigated activated sludge was sampled from the biological treatment tank of the wastewater treatment plant located in Bistrița-RO municipality.

The suspended solids content was determined according to [43]- both by the volumetric (sedimentation for 2 hours) and gravimetric method (drying at 105±3 °C).

Dielectric spectroscopy measurements were performed with specialized equipment AMTEK- 296 Dielectric interface (Solartron Analytical)- both on activated sludge sediment and autoclaved sediment for 30 minutes at 105±3 °C.

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REFERENCES

1. C. Staniloiu; C. Florescu; *Rev. Chim.*, 2014, 65 (4), 502-505.
2. C.M. Dragomir Bâlcănică; C. Munteniţă; A.G. Simionescu; I.G. Bîrsan; *Rev. Chim.*, 2020, 71 (1), 266-272.
3. C. Munteniţa; C.M. Dragomir Balanica; A.G. Simionescu; S. Stanciu; C.L. Popa; *Rev. Chim.*, 2019, 70 (6), 1920-1923.
4. S. Gheorghe; G.G. Vasile; C. Stoica; M. Nita-Lazar; I. Lucaciu; A. Banciu; *Rev. Chim.*, 2016, 67 (8), 1469-1473.
5. D. Cirtina; M.N. Mihut; *Rev. Chim.*, 2020, 71 (2), 315-323.
6. C. Panaitescu; C. Jinescu; A.M. Mares; *Rev. Chim.*, 2016, 67 (5), 925-928.
7. C.M. Dragomir Balanica; A.G. Simionescu; C.L. Popa; C.I. Bîchescu; C. Munteniţa; *Rev. Chim.*, 2019, 70 (5), 1664-1666.
8. M. Mincu; M.I. Marcus; M.A. Mitoi; N.S. Raischi; *Rev. Chim.*, 2018, 69 (12), 3553-3556.
9. I. Lingvay; D.I. Văireanu; K. Öllerer; C. Lingvay; *Environ. Eng. Manag. J.*, 2012, 11 (4), 767-772.
10. N. Groza; A. Manescu; E. Panturu; A. Filcenco-Olteanu; R.I. Panturu; C. Jinescu; *Rev. Chim.*, 2010, 61 (7), 680-684.
11. C. Bumbac; E. Manea; A. Banciu; C. Stoica; I. Ionescu; V. Bădescu; M. Nita-Lazar; *Rev. Chim.*, 2019, 70 (1), 275-277.
12. V.D. Gherman; P. Molnar; M. Motoc; A. Negrea; *Rev. Chim.*, 2018, 69 (4), 806-808.
13. L.I. Dungan; A.E. Ciocablă; V. Pode; *Rev. Chim.*, 2020, 71 (1), 223-227.
14. C.G. Gheorghe; O. Pantea; V. Matei; D. Bombos; A.F. Borcea; *Rev. Chim.*, 2011, 62 (10), 1023-1026.
15. E. Manea; C. Bumbac; A. Banciu; C. Stoica; M. Nita-Lazar; *Rev. Chim.*, 2020, 71 (1), 88-92.
16. E.S. Biris-Dorhoi; M. Tofana; S.M. Chis; C.E. Lupu; T. Negreanu-Pirjol; *Rev. Chim.*, 2018, 69 (5), 1089-1098.
17. D.E. Pascu; C. Modrogan; A.R. Miron; P. C. Albu; D.D. Clej; M. Pascu (Neagu); S. Caprarescu; *Rev. Chim.*, 2015, 66 (12), 1950-1955.
18. I. Lingvay; C. Lingvay; A. Voinea; *Rev. Roum. Sci. Tech. El.*, 2008, 53 (2), 85-94.
19. C. Lingvay; A. Cojocaru; T. Vişan; I. Lingvay; *U.P.B. Sci. Bull. Series B*, 2011, 73 (4), 143-152.
20. I. Lingvay; M. Gabor; C. Lingvay; *Rev. Chim.*, 2006, 57 (2), 180-183.
21. I. Lingvay; C. Lingvay; C. Homan; O. Ciogescu; *Rev. Chim.*, 2006, 57 (12), 1279-1282.
22. I. Lingvay; A.M. Bors; D. Lingvay; L. Radermacher; V. Neagu; *Rev. Chim.*, 2018, 69 (12), 3593-3599.
23. A.M. Bors; N. Butoi; A.R. Caramitcu; V. Marinescu; I. Lingvay; *Mat. Plast.*, 2017, 54 (3), 447-452.
24. A. Caramitcu; N. Butoi; T. Rus; A.M. Luchian; S. Mitrea; *Mat. Plast.*, 2017, 54 (2), 331-337.
25. I. Szatmari; M. Lingvay; L. Tudose; A. Cojocaru; I. Lingvay; Rev. Chim., 2015, 66 (3), 304-311.
26. T. Rus; E. Radu; I. Lingvay; M. Lingvay; O.C. Ciobotea-Barbu; C. Campureanu; F.M. Benga; G.C. Lazar; D.I. Vaireanu; U.P.B. Sci. Bull., 2017, 79 (4), 167-180.
27. J. Filipič; B. Kraigher; B. Tepuš; V. Kokol; I. Mandic-Mulec; Bioresour Technol. 2012, 120, 225-232.
28. R.W. Hunt; A. Zavalinl; A. Bhatnagar; S. Chinnasamy; K.C. Das; Int. J. Mol. Sci., 2009, 10, 4515-4558.
29. B. Lewczuk; G. Redlarski; A. Żak; N. Ziółkowska; B. Przybylska-Gornowicz; M. Krawczuk; BioMed Res. Int., 2014, ID 169459
30. O. Hiwaki; Engineering in Medicine and Biology Society, Proceedings of the 20th Annual International Conference of the IEEE, 1998.
31. Y. Touiltou; B. Selmaoui; Dialogues Clin Neurosci., 2012, 14 (4), 381-399.
32. K. Aronsson; U. Rfnner, E. Borch; Int. J. Food Microbiol., 2005, 99, 19-32.
33. E. Radu; D. Lipcinski; N. Tănase; I. Lingvay; Electroteh. Electron. Autom., 2015, 63 (3), 68-74.
34. C.M. Ferencz; P. Petrovszki; A. Dér; K. Sebők-Nagy; Z. Kóta; T. Páli; Sci. Rep., 2017, 7, 45309.
35. C. Stancu; M. Lingvay; I. Szatmári; I. Lingvay; The 8th Int. Symp. on ATEE, Bucharest, Romania, May 23-25, 2013, 1-4.
36. D. Sandu; I. Lingvay; S. Lányi; D.D. Micu; C.L. Popescu; J. Brem; L.C. Bencze; C. Paizs; Studia UBB Chemia, 2009, 54 (4), 195-201.
37. M. Lingvay; L. Czumbil; Electroteh. Electron. Autom., 2014, 62 (3), 84-89.
38. M. Lingvay; C. Stancu; I. Szatmár; I. Lingvay; Electroteh. Electron. Autom., 2013, 61 (1), 43-47.
39. M. Lingvay; A.R. Caramitu; A.M. Borș; I. Lingvay; Studia UBB Chemia, 2019, 64 (2), 279-288.
40. I.A.A. Al-Darkazly; S.M.R. Hasan; IEEE J. Transl. Eng. Health Med., 2020, 8, 1–13.
41. M. Blank; Electromagn. Biol. Med., 2008, 27(1), 3–23.
42. M. Gao; J. Zhang; H. Feng; Bioelectromagnetics, 2011, 32, 73-78.
43. STAS 6953-81 – Surface water and wastewater. Determination of suspended solids content, calcination loss and calcination residue.
