Gamma Radiation Impact on Agro-industrial Wastewater Treatment

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Abstract

Agro-industrial wastewaters have high quantity of recalcitrant compounds which blocks biological treatment. With the objective of optimization of these processes the impact of ionizing radiation on swine and dairy wastewater was undertaken. The irradiations were performed in a semi industrial Co-60 source at a dose rate of 0.5 kGy/h using absorbed doses from 5 up to 30 kGy. For these studies, the main wastewater quality parameters, such as Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), were determined. Conventional microbiological techniques were used to assess microbial gamma radiation inactivation. After irradiation, the swine wastewater analysis has shown a decrease of the TSS values. In the opposite an increase of the COD is presented. In the case of the dairy wastewater the same phenomena is observed. However, for the dairy wastewater there is a decrease of BOD with dose which could lead to a better water quality. Results on the microorganisms' response to gamma radiation were analyzed in terms of the D-value. For the swine wastewater the D-value was 1.4 kGy, and for the dairy wastewater the D-value was 2.7 kGy. The overall results point out to the advantages on the use of ionizing radiation being a useful tool as complement of conventional treatment.

Keywords: gamma radiation, dairy wastewater, swine wastewater, organic matter, scissor effect, microorganisms

Introduction

In the last few years, research on the application of new technologies for advanced wastewater treatment has been widely developed [1]-[4]. For the toxic organic matter removal which is frequently present in wastewater, like those generated in different types of agro-industrial plants, biological treatment (e.g. bioreactors, anaerobic/aerobic lagoons) seems to be a less costly alternative [5], [6]. However, some disadvantages are found in this kind of wastewater due to the presence of biorecalcitrant compounds that blocks the biological processes [7]. Therefore, it is crucial to understand how those biorecalcitrant compounds can be eliminated or transformed in order to turn biological processes feasible. Chemical treatment is also often carried out for the elimination of hazardous organic pollutants, by means of precipitation agents and flocculation [8]. However, the addition of chemicals induces the increase of pollutants in the wastewater. Advanced oxidation processes (AOP) have been studied with the objective to reduce the concentration of wastewater pollutants [9], [10], [11]. Oxidation of organic matter, namely by ionising radiation, leads to the decrease of wastewater quality parameters, such as Chemical Oxygen Demand (COD) and Biochemical Oxygen Demand (BOD) [12], [13]. In this study, the application of gamma radiation as AOP in dairy and swine wastewater was studied. Dairy wastewater

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have high concentration of proteins and whey due to the milk and derivatives production and the swine wastewater is characterized by high concentration of phosphates and nitrates which blocks the biological treatment by eutrophication of the system [14]. Therefore, the effects of gamma irradiation on the main wastewater quality parameters were analysed in order to predict the implementation of such technologies to upgrade the wastewater treatment yields in industry.

Materials and Methods

Irradiation

The effluent used for the study was untreated wastewater from dairy and swine industry which were submitted to mechanical pre-treatment screening only. Samples in a 0.5 L beaker were irradiated at gamma radiation and the local dose rate was previously determined by reference Fricke dosimeter [15]. The irradiation doses were established from 5 up to 35 kGy at dose rate 0.5 kGy h⁻¹ and were measured by routine dosimeters (Harwell Red Perspex, Batch HA Type 4034; and Gammachrome YR), with nominal uncertainty limits of about 5% [16].

Analytical methods

BOD (Method 5210 B, Aqualytic BOD-OxiDirect), COD (Tritimetric Method 5220 C, Merck Thermoreactor TR300) and TSS (Method 2540 B) were measured according to Standard Methods for the Examination of Water and Wastewater [17]. COD was measured after filtration with membrane pore size 1.2 μ m. All values are the average of 3 replicates. The impact of gamma radiation on the mesophilic wastewater natural contaminants was determined by means of total counts before and after irradiation. The method was based on the

Spread Plate Method (9215 C) as described in the Standard Methods for the Examination of Water and Wastewater [17]. For each sample, two dilutions were performed and three replicas of each dilution were plated in Tryptic Soy Agar (TSA, Oxoid, UK). The growth conditions were aerobic incubation at 30 °C for 14 days. Colony forming units (cfu) were counted after 24, 48 and 72 h after 7 and 14 days. The inactivation response of natural wastewater microbiota was assessed by the D-values parameter determined based on the survival curves. Inactivation efficiency was calculated according to the equation Efficiency (%)=[N0-Nd)/N0]*100. Where Nd, is the number of survivors after irradiation at several doses and N0 the initial count (non-irradiated sample). Survivor isolates were morphologically characterized (macroscopic, microscopic and biochemically).

Results

The effect of gamma radiation on COD and TSS values is different for the two kinds of wastewater. The COD and the TSS were measured in the irradiated and non irradiated wastewater after filtration with a 1.2 μ m filters (Fig. 1 and 2). The COD-values are the average of 3 replicates (α = 0.05).

As shown in Fig. 1, there is a decrease of the TSS values after irradiation. In the



Fig. 1: COD (n=15; a=0.05) and TSS (n=5; standard error: 10%) versus absorbed dose (dose rate = 0.5 kGy/h) for swine wastewater.

opposite an increase of the COD values is presented. These phenomena could be explained due to the increase of species in solution (decrease of TSS) that leads to a higher COD parameter at the same values of absorbed dose.

In the case of the dairy wastewater the same phenomena is observed as shown in Fig. 2, which also could be explained by radiation-induced scissor effect that degraded the organic pollutants and there are an increase of chemical species soluble in the water. The microbiological population and the chemical species are dynamically inter-dependent, thus studies on wastewater treatment need multidisciplinary analysis in order to fully understand them. In the Fig. 3 and Fig. 4 it is presented the relation between the BOD values and the survival microorganisms.



Fig. 2: COD (n=15; a=0.05) and TSS (n=5; standard error: 10%) versus absorbed dose (dose rate = 0.5 \text{ kGy/h}) for dairy wastewater.

The reduction of BOD is related to the biodegradation of organic matter [18], therefore results point out to a better water quality (the higher the dose absorbed, the lower the BOD value). As shown in the Fig. 3 there is a decrease of the BOD values and the number of the total microorganisms which means there is an increase of the water quality.



Fig. 3: BOD (n=5; standard error= 10%) and survival curve ($3 \le n \le 9$; a = 0.05) versus absorbed dose in the dairy wastewater sample.



Fig. 4: BOD (n=5; standard error= 10%) and survival curve ($3 \le n \le 9$; a = 0.05) versus absorbed dose in the swine wastewater sample.

In swine wastewater BOD parameter shows the high level of the organic matter and is consistent with the results of COD (Fig. 1). The connection between microbiota and the parameter BOD could be explained by different hypotheses namely by the microorganisms' radiosensitivity. Although scissor effect was shown (higher values of BOD) microbiota data point out to an inability to use the by-products of scissor effects.

The effects of gamma radiation on natural microbiota wastewater samples were evaluated by means of D-value parameter and inactivation efficiency for each absorbed dose (TABLE 1). D-value was determined based on the inverse of the slope obtained by regression analysis of the linear kinetic inactivation curve (n=15) (Fig. 3 and Fig. 4)

Table 1:Wastewater mesophilic microbiota D-value and inactivation efficiency to gamma radiation at 0.5 kGy/h.

Sample	Dose	D-	Efficiency
	(kGy)	value	(%)
Dairy Wastewater	4.82	2.7	99.76
	9.00		99.96
	15.39		99.59
	25.41		99.83
Swine Wastewater	5.21	1.3	99.99
	7.50		>99.99
	20.13		>99.99
	31.76		>99.99

Microbiota from swine wastewater showed the highest sensitivity to ionising radiation in the studied circumstances (lowest D-value). This result was in conformity with the obtained microbial survivors' morphological characterization. The most resistant morphological types isolated from 30 kGy irradiated samples were gram negative cocci (42%) and gram negative rods (50%) from dairy and swine wastewater, respectively. As recognized [19], the gram negative rods indicated to be less radiation resistant comparatively to cocci. The substrata and the type of contaminants (natural microbiota) are determinant factors for the response to the lethal agent as can be observed in the D-value and efficiencies obtained for the two types of wastewater.

Conclusions

Based on these results, the combining ionising radiation with a conventional treatment process could lead to better yields in the treatment of the agro-industry wastewater. Results showed that lower dose rate (0.5 kGy.h⁻¹) can lead to better bioremediation processes in both wastewater substrates due to the irradiation effect on its degradation, making molecular forms easier to be metabolized by microorganisms, consequently reducing the wastewater chemical pollution and possibly the residence time in the lagoons.

In the future, it is intended to implement a dynamic process that permits to be step ahead in getting a range of physical parameters connected with the biological and chemical responses for a technical and economical benefits design.

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References

- A.L. Filby, J.A: Shears, B.E. Drage, J.H. Churchley, C.R. Tyler CR, "Effects of Advanced Treatments of Wastewater Effluents on Estrogenic and Reproductive Health Impacts in Fish", Environmental Science & Technology, vol. 44, pp. 4348-4354, Jun. 2010.
- D.M. Kargbo, "Biodiesel Production from Municipal Sewage Sludges", Energy & Fuels, vol. 24, pp. 2791-2794, May 2010.
- 3. N. Li, H.Z. Ma, B. Wang B, "Advanced treatment of paper mill wastewater by catalytic vacuum distillation", Desalination and Water Treatment, vol. 16, pp. 298-303, Apr. 2010.

- L. Wojnárovits, E. Takács, "Irradiation treatment of azo dye containing wastewater: An overview", Radiat. Phys. Chem., vol. 77, pp. 225-244.
- A. Akcil, A. G. Karahan, H. Ciftci, O. Sagdic, "Biological treatment of cyanide by natural isolated bacteria (Pseudomonas sp.)", Minerals Engineering, vol. 16, pp. 643-649, Jul. 2003.
- M. D. Knoblock, P. M. Sutton, P. N. Mishra, K. Gupta, A. Janson, "Membrane Biological Reactor System for Treatment of Oily Wastewaters", Water Environment Research, Vol. 66, pp. 133-139, Apr. 1994.
- C.A. Fewson, "Biodegradation of xenobiotic and other persistent compounds: the causes of recalcitrance", Trends in Biotechnology, vol. 6, pp. 148-153, Jul. 1988.
- 8. J. Bratby, Coagulation and Flocculation in Water and Wastewater Treatment, 2nd ed, London, IWA Publishing, 2006.
- F. L. Rosario-Ortiz, E.C. Wert, S.A. Snyder SA, "Evaluation of UV/H2O2 treatment for the oxidation of pharmaceuticals in wastewater", Water Research, vol. 44, pp. 1440-1448, Mar. 2010.
- J. Tuerk, B. Sayde, A. Boergers, H. Vitz, T.K. Kiffmeyer, S. Kabasci, "Efficiency, costs and benefits of AOPs for removal of pharmaceuticals from the water cycle", Water Science and Technology, vol. 61, pp. 985-993, 2010.
- S. Yao, D. Dou, H. Fu, S. Liu, S. Wang, X. Sun, "Innovation technique of radiation for the treatment of 4-chlorphenol as a model of POPs in waste water", Nuclear Instruments

and Methods in Physics Research B, vol. 236, pp. 266-271, May 2005.

- R. Melo, S. Cabo Verde, J. Branco, M.L. Botelho, "Gamma radiation induced effects on slaughterhouse wastewater treatment", Radiat. Phys. Chem., vol. 77, pp. 98–100, 2008.
- K. Kubesch; R. Zona; S. Solar; P. Gehringer, "Ozone, Electron Beam and Ozone-Electron Beam Degradation of Phenol. A Comparative Study", Ozone: Science & Engineering, vol. 25, pp. 377-382, Oct. 2003.
- 14. J. K. Cronk, "Constructed wetlands to treat wastewater from dairy and swine operations: a review", Agriculture, Ecosystems and Environment, vol. 58, pp. 97-114, Dec. 1996.
- 15. K. Sehested, The Fricke Dosimeter, N. H. Berry Ed., New York, Marcel Dekker, 1970.
- McLaughlin, W.L. Boyd, A.W., Chadwick, K.H., McDonald, J.C., Miller, A., Dosimetry for radiation processes, London, Taylor & Francis Eds. 1989.
- American Public Health Association, The Standard Methods for the Examination of Water and Wastewater, 20th ed, Washington DC, American Public Health Association, 1998.
- T. Wang, T.D. Waite, C. Kurucz, W.J. Cooper, "Oxidant reduction and biodegradability improvement of paper mill effluent by irradiation", Wat. Res., vol. 28, pp. 237-241, 1994.
- 19. BLOCK, S.S., "Disinfection, Sterilization and Preservation", Lea & Febiger, Philadelphia, Third Edition (1983) 27–33.