CHEMICAL ENGINEERING TRANSACTIONS

VOL. 96, 2022

A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it

Guest Editors: David Bogle, Flavio Manenti, Piero Salatino

ISBN 978-88-95608-95-2; **ISSN** 2283-9216

DOI: 10.3303/CET2296018

Coupling the Infiltration-Percolation and Solar Distillation Technologies for the Treatment of Olive Mill Wastewater

Soufiane Bakri Alaoui[a,b], Mounia Achak[b,c], Younes Chhiti[d], Fatima Ezzahra[e], M'Hamdi Alaoui[e], Edvina Lamy[a,*]

[a]University of Technology of Compiègne, Alliance Sorbonne University, Department of Chemical Enginneering, UTC/ESCOM, EA 4297 TIMR, Centre de recherche Royallieu - CS 60 319 - 60 203 Compiègne cedex, France
[b]National School of Applied Sciences, Chouaïb Doukkali University, (LabSIPE), El Jadida, Morocco
[c]Mohammed VI Polytechnic University (UM6P), Chemical and Biochemical Sciences, Benguerir, Morocco
[d]National school of chemistry, University Ibn Tofail, Kénitra, Morocco
[e]Sciences Faculty, University Abdelmalek Essaâdi,Tétouan, Morocco
edvina.lamy@utc.fr

The current study was carried out to treat the Olive Mill Wastewater (OMW) via a hybrid system that combine the infiltration-percolation system using low-cost natural adsorbents and solar distillation technology. The experimental pilot was composed of an aerated PVC column filled with an alternation of a permeable layer of pouzzolane and a mixed layer of sand and natural low-cost adsorbents as charcoal and sawdust. After infiltration-percolation experiments, the recovered filtrate was inserted to the solar still to continue the treatment. The Higher Heating Value (HHV) of the solid residue after solid still treatment was investigated in this work. The results showed a pollutant removal rate by infiltration-percolation technique of 73.9 % of Chemical Oxygen Demand (COD) and 84.1 % of polyphenols. The OMW treatment efficiency was improved after treating the filtrate by solar distillation technology. Indeed, the removal rate increased to reach 98.8 % and 99.2 % for COD and polyphenols respectively and a colorless and transparent distillate was recovered. The HHV of the solid residue that remained in the basin of the solar still was 16.8 MJ/kg, which makes it a valuable source of biofuel.

1. Introduction

Olive Mill Wastewater (OMW) is a liquid by-product generated during the olive oil production which represents a serious environmental problem for the Mediterranean countries especially, since they are responsible of the production of 98 % of the entire worldwide olive oil (Gullón et al., 2020). This effluent is characterized by a high load of organic matter with a Biological Oxygen Demand value (BOD5) of 15–135 g/L) and Chemical Oxygen Demand (COD) of 37–318 g/L (Khoufi et al., 2015). OMW contains also a high load of phenolic compounds that range between 1 and 25 g/L and an acid pH, which confer to this effluent an anti-microbial effect rending natural biological degradation of contaminants in water much harder if it's thrown into waterways. This could lead to a suppression of germination and reduce the growth of the plant's root (El Hassani et al., 2009).
Different technologies have been used for OMW treatment. One of them include the infiltration-percolation technique that combines physical, chemical and biological processes. When the contaminated effluent percolate through a filter bed, particulate organic matter is removed by surface filtration and adsorption phenomena (Achak et al., 2011). Solar distillation technology, a technology mostly used for the desalination of seawater (Abujazar et al., 2016) has also been applied for treating OMW as well (Jaradat et al., 2018). This technology uses the solar radiation to evaporate the wastewater, reducing its original volume and rendering the remaining sludge easier to manage for further treatment. Its principle is based on the creation of the greenhouse effect where the incident solar energy passing through a transparent cover, is absorbed by a black surface, which heats up and emits in the field of long wavelengths. These rays can no longer pass through the glass, thus creating a heat trap (Boukerzaza et *al*., 2007).

Paper Received: 23 December 2021; Revised: 11 July 2022; Accepted: 20 September 2022
Please cite this article as: Bakri Alaoui S., Achak M., Chhiti Y., M'Hamdi Alaoui F.E., Lamy E., 2022, Coupling the Infiltration-percolation and Solar Distillation Technologies for the Treatment of Olive Mill Wastewater, Chemical Engineering Transactions, 96, 103-108 DOI:10.3303/CET2296018

At the best of our knowledge, the treatment of OMW using a hybrid system combining both technologies of infiltration-percolation and solar distillation has not been reported in the literature, It is known that the infiltration-percolation is an efficient technology to treat complex and heterogenous pollutant effluents but the solar distillation technology could not only improve pollutant removal rate but it could allow energetic valorization of OMW effluent residues after treatment as well. For this reason, the main goals of this work were (i) to identify the performance of treatment of OMW by infiltration-percolation using a multi sand layering with natural and low-cost adsorbents such as charcoal, sawdust and pouzzolane, (ii) to compare the performance of each technology separately and the hybrid system in treating OMW, (iii) to evaluate energetically the solid residue in the solar still.

2. Materials and methods

2.1 Experimental devices

The treatment process was conducted using two experimental pilots. The first one consisted of an opaque PVC column with 10 cm in diameter and 110 cm in height (Figure 1). The column was filled with different layers: a 10 cm gravel layer at the bottom as a drainage layer, followed by an alternation of mixing layer and a permeable layer. The mixed layer contains 70 % of sand, 20 % of charcoal and 10 % of sawdust with a grain size ranged between 0.3 and 2.5 mm while the permeable layer was constituted by 100 % of coarse particles of pouzzolane with a grain size ranging from 2.5 to 5 mm. To enhance the organic matter degradation, the column was equipped with different air entries, consisting in lateral small columns of 3 cm in diameter, ensuring air penetration to the center of the filter bed and promoting a good condition for microbial degradation of the organic matter.
After the infiltration-percolation experiments, the solar distillation process was performed in an experimental device, consisting on a 2,800 cm² iron sheet basin whose interiors walls were painted in black to absorb the maximum of solar radiation, thus increasing the distillation rate of the OMW. The top of the device was covered by a glass cover with a 30° slope and a gutter that led to a storage bottle to collect the distillate was attached to the bottom of the glass. The edges of the glass were sealed to make the entire still airtight (Figure 1).
The solar still unit was placed outdoor, faced toward the north-east to catch the maximum solar radiation and increase the temperature inside the solar system by creating a greenhouse effect. The basin was filled with OMW collected after the infiltration-percolation process, and to test the efficiency of the solar still separately, a distillation experiments have been carried out with the raw OMW as well.

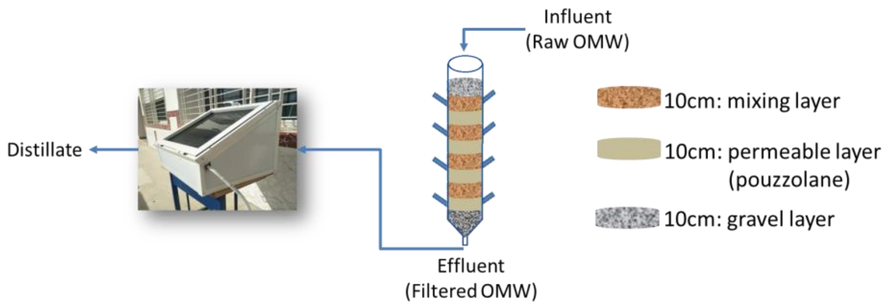

Figure 1: Schematic representation of the infiltration percolation device and actual solar still picture

2.2 Physical chemical analysis of OMW

Both influents and effluents were analyzed to obtain OMW physical-chemical characterization. pH and electrical conductivity (EC) were continuously measured by using a pH meter (WTW) and a conductivity meter 950 respectively. The total Chemical Oxygen Demand (COD) was determined by colorimetric methods (APHA 1992): the sample was introduced into a digestion solution and the mixture was then incubated at 150 °C in a COD reactor for 2h. Then, the COD concentration was deduced by absorbance measurements using UV/VIS spectrophotometer (JENWAY 63200D) at a wavelength of 620 nm. Phenolic compounds were quantified by the Folin-Ciocalteu method (Box, 1983), using caffeic acid as standard and the absorbance was determined at 765 nm by using a UV-visible spectrophotometer.
Ammonia nitrogen (NH_4^+-N) in raw OMW was measured by the distillation method (Kjeldahl method). Nitrate (NO_3^--N) was determined by the method of salicylate (Monteiro et *al*., 2003), which is a colorimetric method and the concentration of Nitrate was deduced by absorbance measurement at 415 nm. The phosphorus content was

determined by a spectrophotometry method according to AFNOR (T90-023). The percentage of removal, R, for each parameter that represents the treatment efficiency of the column was calculated as following:

$$\boldsymbol{R} = ((\boldsymbol{Ci} - \boldsymbol{Ce})/\boldsymbol{Ci}) * \mathbf{100} \quad (1)$$

where Ci and Ce are the concentration of the influent and the effluent respectively.

2.3 Higher Heating Value (HHV) measurement and elementary analysis

The Higher Heating Value (HHV) measurement of solid residue that remain in the basin after the solar distillation process was determined using the calorimetric bomb "IKA C2000 Calorimeter Basic" according to the standard method ISO-1928. A crucible that contains the sample was inserted into the bomb, which was closed and filled with oxygen under a pressure of 30 bars. The bomb was then placed in the adiabatic calorimetric equipment filled with water and the sample ignited electrically. The resulting increase of the water temperature allows the calculation of the HHV of the sample. The heat capacity of the calorimeter was determined using benzoic acid as reference substance. The elementary analysis of this residue was determined on a dry sample using CHNS-O Flash 2000 which is an organic elemental analyzer.

3. Results and discussion

3.1 OMW characterization

Mean values of triplicate measurements of each physical-chemical parameter of raw OMW as well as adsorbents characteristics are summarized at Table 1. OMW was characterized by a high degree of acidity with a pH of 5.15, mainly due to the high load of organic acid such as phenolic acid and fatty acid (Elabdouni et *al*., 2020), because during the storage period, auto-oxidation and polymerization reactions transform phenolic alcohols into phenolic acids. These reactions are manifested by a change in the initial color of OMW from brown reddish toward a dark black color.

The high conductivity values of OMW (12.8 mS/cm) was due both to the high richness of OMW of mineral salts as well as the salting process used for olive preservation (Elabdouni et *al*., 2020). The OMW was characterized by a high load of organic matter with a total and dissolved COD value of 136,22 gO_2/L and 68.51 gO_2/L respectively. The phenolic compounds concentration of 1.38 g/L conferred to OMW effluent an aspect of toxicity (Elabdouni et *al*., 2020). High concentration values of mineral content were obtained in the OMW (Table 1).

Table 1: Physical-chemical characterization of raw OMW and mixed sand and pouzzolane

	Raw OMW	Mixed sand	Pouzzolane
pH	5.15	8.83	6.72
Conductivity (mS/cm)	12.8	1.52	0.499
Total COD (gO_2/L)	136,22±1.56		
Dissolved COD (gO_2/L)	68.51±3.15		
Total phenols (g/L)	1.38±0.09		
NH_4^+ (mg/L)	7.51±0.72		
NO_3^- (mg/L)	0.13±0.01	0.23	ND
PO_4^{3-} (g/L)	0.26±0.01		
Total P (g/L)	0.68±0.03		
Total solid (g/g)		0.9949	0.98
Organic matter (mg/g)		41	254
Mineral matter (g/g)		0.997	0.999
Apparent density (g/cm^3)		1.19	0,855
Reel density (g/cm^3)		2.30	2,548
Porosity (%)		48.26	65.26

3.2 Treatment efficiency of infiltration-percolation technology

The pH value of raw OMW at the inlet of the column was acid 5.36. After the infiltration-percolation the pH increased to the neutrality and become 7.38 (Figure 2.a). These results are in accordance with those previously reported by Achak et *al*., (2009). This behavior could be due to the fixation of organic acids, responsible for the low pH value, in the medium filter particles. Adsorption and ion exchange are the principal physical-chemical surface reactions that occur on this medium during the transfer of wastewater through it (Achak et *al.*, 2019). According to these authors, this behavior could be explained by the dilution of the carbonate salts from the porous media or because of the biological oxidation as well. Besides that, the soil and the pouzzolane are known by their buffer capacity that explain the variation of the pH from the acidity to the neutrality (Achak et *al*., 2019).

The value of EC increased after passing through the bed filter from 12.8 mS/cm to 17.4 mS/cm (Figure 2.b). This increase could be attributed to the scrubbing of sand minerals and the mineralization of the organic matter and these results were in agreement with those performed by Achak et *al*., (2019).

COD is one of the most important parameters that represent the load of organic matter in the liquid effluent and also reflect the treatment efficiency of each technology. In this study the infiltration-percolation technique showed a removal rate of 73.9 % for COD (Figure 3). This rate corresponded to a concentration of 35.3 gO_2/L at the outlet of the column. The results obtained are similar to these found by Achak et *al*., (2011); Arafa et *al*., (2018) and they could be explained by the elimination of organic matter by physical processes such as sedimentation, adsorption and particle filtration. In addition, the presence of pozzolane substrate could also participate in the removal of the organic matter by adsorption, as its adsorption capacity regarding organic matter was demonstrated by Siéliéchi et *al*., (2012).

The polyphenols concentration at the outlet of the column reached 0.22 g/L, which corresponded to a removal rate of 84.1 % (Figure 3). This percentage was among the highest removal rate of polyphenols that has been recorded for treating raw OMW compared to that found by (Achak et *al.*, 2009; Arafa et *al*., 2018). It should be noted that the phenolic compounds degradation by bacteria may be accelerated by a slight rise in the pH and/or an improvement of ventilation, which corresponded exactly to the experimental conditions of this study. In addition, the neutralization of the pH transforms phenols into phenolates, that are well retained by the iron and aluminum oxides, calcium carbonates and silicates of the porous materials (Achak et *al*., 2009).

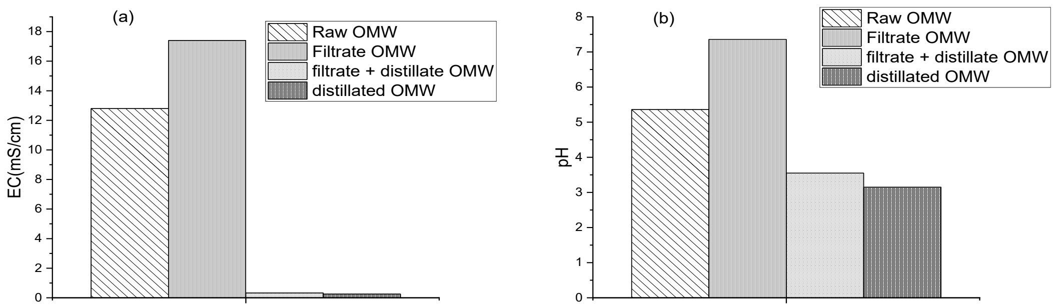

Figure 2: The EC (a) and the pH (b) of the raw OMW; filtrate OMW; filtrate and distillated OMW; and distillated OMW

3.3 Treatment efficiency of raw OMW by solar distillation technology

The treatment of raw OMW with the solar still, lead to obtain a colorless distillate which means that the compounds responsible for the OMW's color has been eliminated. The physical chemical measurements showed that the recovered distillate was characterized with an acid pH (3.15) (Figure 2.a) and a decrease of EC from 12.8 mS/cm to 252 µS/cm was also observed (Figure 2.b). For the polyphenols, the solar still contributed to a removal rate that reached 98.78 % (Figure 3), that corresponds to a concentration of 0.016 g/L. And 92.34 % of the organic matter has been removed, with a recovered distillate that has 10.62 gO_2/L, which was due to the transfer of volatile organic matter.

3.4 Treatment efficiency improvement by the hybrid system

The hybrid system consisted in introducing the filtrate OMW recuperated from the infiltration-percolation to the solar still in order to improve the efficiency of the treatment and remove the maximum of the polluting matter.

The results showed that the pH and the EC decreased from 5.15 and 12.8mS/cm to 3.55 and 329 µS/cm respectively (Figure 2). The removal rate of polyphenols reached 99.2 %, corresponding to a concentration of 0.01 g/L, which is almost the same concentration observed while using the solar still only (Figure 3). This concentration is slightly lower than that reported by Jaradat et *al*., (2018). According to (Jaradat et *al*., 2018), the remaining part of total phenols in the basin or part of it is either being sorbed to the remained solid fraction or being degraded photochemically especially in the presence of infrared radiation that passes through the glass cover in solar still. As reported by Zhou et al., (2016), the higher temperature and infrared radiation accelerate the degradation rate of total phenolic compounds. However, for the organic matter, the system gave a removal rate of 98.8 %, with a recovered distillate with a COD of 1.6 gO_2/L.

This concentration is 6.6 times lower that that observed while using the solar still only, which proved that coupling the infiltration-percolation and the solar distillation gives the best results in removing the pollutant matter.

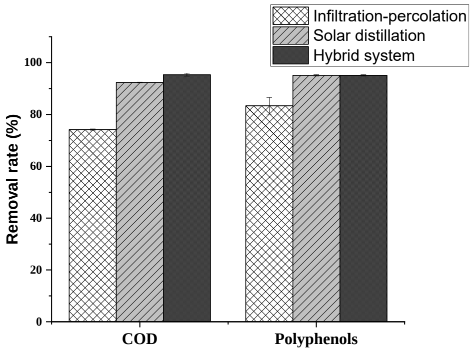

Figure 3: Removal rate of COD and polyphenols after treating OMW by infiltration-percolation technology, solar still technology and hybrid system (combination of both technologies) relative to raw OMW

3.5 Energetic valorization of the residue

After drying the solid residue remained in the basin after solar distillation treatment, the higher heating value was calculated after the equation, established by Channiwala and Parikh, (2002), that relates the percentage of C, H, N, S, O and ash, based on the elementary analysis of solid residue (Table 2), with HHV value :

$$\boldsymbol{HHV = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.0151N - 0.0211A} \ (MJ/kg) \quad (2)$$

Where 0% ≤ C ≤ 92.25%, 0.43% ≤ H ≤ 25.15%, 0.00% ≤ O ≤ 50.00%,0.00% ≤ N ≤ 5.60%, 0.00% ≤ S ≤ 94.08%, 0.00% ≤ A ≤ 71.4%, 4.745MJ/kg ≤ HHV ≤ 55.345 MJ/kg where, C, H, O, N, S and A represents carbon, hydrogen, oxygen, nitrogen, sulphur and ash contents of the material, respectively, expressed in mass percentages on dry basis. According to this equation the theoretical HHV reached 17.34 MJ/kg and it was similar to the experimental value.

Table 2: Calorific value and elementary analysis of solid residue

Number of samples	HHV (Mj/kg) (dry basis)	Moisture (%)	Ash (%) (dry basis)	Elementary analysis (dry basis)				
				C (%)	H (%)	N (%)	S (%)	O (%)
3	16.8±0.44	7.48±0.37	19.51±0.96	42.6±2.11	5.2±0.54	0.89±0.17	0.3±0.03	31.5±0.11

The results showed that this residue had a considerable HHV of 16.8 MJ/kg (Table 2), which was higher than that of rice straw, millet grain waste (Channiwala and Parikh, 2002), and it was in the same range as that obtained for dry firewood (World Nuclear Association).These results showed that the solid residue left in the solar still after treatment can actually be used as a valuable source of biofuel.

4. Conclusion

This study showed that the infiltration percolation is an efficient technology to treat complex and heterogenous pollutant effluents such as the OMW, leading to an important pollutant removal rate in terms of COD and polyphenols. On the other hand, the solar distillation technology showed a better removal rate for all the parameters compared to infiltration-percolation technology and also eliminated the dark color of the OMW leading to recuperate a colorless distillate, that could be used for watering plants after the neutralization of its pH. Moreover, the use of the hybrid system which was a combination of infiltration-percolation and solar distillation technology ensure the best pollutant removal rate. In addition, the solid residue that remained in the solar still had a considerable HHV of 16.8 MJ/kJ which means that it can actually be used as a valuable source of biofuel. In conclusion, the use of low-cost and eco-friendly adsorbents in infiltration-percolation technology and possible valorization of residues issued by solar distillation technology application are encouraging results that support the idea of circular economy and zero waste technology concept.

Acknowledgments

This work is done as a part of a PhD thesis benefiting from the funding of the region Hauts-de-France and National Center for Scientific and Technical Research CNRST-Morocco.

References

Abujazar, M.S.S., Fatihah, S., Rakmi, A.R., Shahrom, M.Z., 2016. The effects of design parameters on productivity performance of a solar still for seawater desalination: A review. Desalination 385, 178–193. https://doi.org/10.1016/j.desal.2016.02.025

Achak, M., Boumya, W., Ouazzani, N., Mandi, L., 2019. Preliminary evaluation of constructed wetlands for nutrients removal from olive mill wastewater (OMW) after passing through a sand filter. Ecological Engineering 136, 141–151.

Achak, M., Ouazzani, N., Mandi, L., 2009. Treatment of modern olive mill effluent by infiltration-percolation on a sand filter. Revue des sciences de l'eau/Journal of Water Science 22 (3): 421-33 (in French)

Achak, M., Ouazzani, N., Mandi, L., 2011. Organic pollutants removal from olive mill wastewater by a combined system of a sand filter and an aquatic plant system. Revue des sciences de l'eau/Journal of Water Science 24 (1): 35-51. (in French).

Arafa, A.-H., Ouazzani, N., Latrach, L., Hejjaj, A., Assabbane, A., Belkouadssi, M., Mandi, L., 2018. Feasibility of Olive Mill Wastewater treatment by Multi-Soil-Layering Ecotechnology. Journal of Materials and Environmental Sciences 9, 1223–1233.

Boukerzaza, N., Chaker, A., Haddad, Z., 2007. Influence of global irradiation on characteristics operation of a solar still, Revue des Energies Renouvelables ICRESD-07 229 – 234. (in French)

Box, J.D., 1983. Investigation of the Folin-Ciocalteau phenol reagent for the determination of polyphenolic substances in natural waters. Water research 17, 511–525.

Channiwala, S.A., Parikh, P.P., 2002. A unified correlation for estimating HHV of solid, liquid and gaseous fuels. Fuel 81, 1051–1063.

El Hassani, F.Z., Zinedine, A., Amraoui, M.B., Errachidi, F., Alaoui, S.M., Aissam, H., Merzouki, M., Benlemlih, M., 2009. Characterization of the harmful effect of olive mill wastewater on spearmint. Journal of hazardous materials 170, 779–785.

Elabdouni, A., Haboubi, K., Merimi, I., El Youbi, M.S.M., 2020. Olive mill wastewater (OMW) production in the province of Al-Hoceima (Morocco) and their physico-chemical characterization by mill types. Materials Today: Proceedings 27, 3145–3150.

Gullón, P., Gullón, B., Astray, G., Carpena, M., Fraga-Corral, M., Prieto, M.A., Simal-Gandara, J., 2020. Valorization of by-products from olive oil industry and added-value applications for innovative functional foods. Food Research International 137, 109683.

Heat values of various fuels - World Nuclear Association [WWW Document], n.d. URL https://www.world-nuclear.org/information-library/facts-and-figures/heat-values-of-various-fuels.aspx (accessed 2.10.22).

Jaradat, A.Q., Gharaibeh, S., Abu Irjei, M., 2018. The application of solar distillation technique as a mean for olive mill wastewater management: Application of solar distillation technique. Water and Environment Journal 32, 134–140.

Khoufi, S., Louhichi, A., Sayadi, S., 2015. Optimization of anaerobic co-digestion of olive mill wastewater and liquid poultry manure in batch condition and semi-continuous jet-loop reactor. Bioresource Technology 182, 67–74.

Monteiro, M.I.C., Ferreira, F.N., de Oliveira, N.M.M., Ávila, A.K., 2003. Simplified version of the sodium salicylate method for analysis of nitrate in drinking waters. Analytica Chimica Acta 477, 125–129.

Siéliéchi, J.M., Lartiges, B.S., Ndi, S.K., Kamga, R., Kayem, G.J., 2012. Mobilisation of Heavy Metals from Natural Pozzolan by Humic Acid: Implications for Water and Environment. International Journal of Environmental Protection 2, 5.

Zhou, L., Cao, Z., Bi, J., Yi, J., Chen, Q., Wu, X., Zhou, M., 2016. Degradation kinetics of total phenolic compounds, capsaicinoids and antioxidant activity in red pepper during hot air and infrared drying process. International Journal of Food Science & Technology 51, 842–853.