Study of the Transfer of ^{238}U and ^{232}Th During the Treatment of Urban Wastewaters by Infiltration Percolation Through a Phosphate Bed

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Abstract

Naturally occurring radionuclides of terrestrial origin exist ubiquitously in the environment and contribute significantly to external and internal doses to the population. Among them, of particular importance are the solid radionuclides belonging to the ^{238}U and ^{232}Th series which are present in soil and rocks. The aim of this work is to study the transfer of ^{238}U and ^{232}Th during the treatment of waste water by infiltration percolation through a phosphate bed. To assess radiation dose due to the treated water which is used in agriculture and for the irrigation of gardens in urban areas, these radionuclides have been measured in waste water, phosphate samples and treated water by using solid state nuclear track detectors.

Key words: *Treatment, wastewater, radioactivity, raw phosphate, SSNTD, Thorium, Uranium.*

1. Introduction

In Morocco, the mining areas are located in places with a low water potential. The use of unconventional water, such as the reuse of treated wastewater, for the washing of phosphates could meet the demand for water which still growing. This anchors the recycling and reuse of wastewater in the culture of the environment which is part of a sustainable development vision.

The contribution to the recycling of urban wastewater was conducted using the process of infiltration percolation based on a well-defined size of a phosphate bracket. The study has shown that the new bracket used has interesting performances in terms of removing the suspended particles and organic pollution by infiltration, percolation, adsorption **[1]**, chemical reaction, biological pathway, predation and absorption mechanisms of plants or purification process **[2-4]**.

Treatment systems in the soil are able to achieve the elimination levels comparable to the best technologies of wastewater treatment, with additional benefits such as recovery and reuse of water and nutrients for crop production **[5, 6]**.

Studies in the field and in the laboratory led obtaining excellent processing performance of domestic wastewater by infiltration **[7]**. With removal rates of COD, suspended solids, and the viruses were more than 80% of phosphorus removals of almost 100% **[1, 8-12]**.

The radioactivity of phosphates has always been a concern, because it has uranium levels between 100 and 200 ppm depending on the site **[13]**. A study of the distribution of radioactivity, especially for uranium and thorium, in the various components of the filtration bed is required to check whether the radioactivity exceeds the international standards set at the treated water. In this context, we used the CR-39 and LR-115 type II solid state nuclear track detectors (SSNTDs).

2. Materials and methods

a. Raw phosphate

The material used in this study is the phosphate of Khouribga city. To remove impurities, the media was thoroughly washed with distilled water. This process was repeated several times until the removal of impurities **[14]**, and then dried in an oven at 105 °C.

Sample was characterized by X-ray diffraction using a Siemens D500 powder diffractometer. The scanning electron microscope SEM (Zeiss DSM950) was used to determine the morphology of materials. The specific surface area was determined by the BET method using a Micromeritics ASAP2420. The size of the porous media was determined by the laser particle size Cilas. Table 1 summarizes the results:

Table 1 : Characterization of draining materials

Settings	Phosphate
Dm (Åm)	238,30
D10 (Åm)	136,86
D90 (Åm)	358,61
Uniformity coefficient	2
SEM	irregular or rounded grains
Surface area (m^2/g)	12,74

Analysis by DRX indicates that the natural phosphates include more than 200 mineral species, but the most common belong to the family of apatite. The phosphate minerals of sedimentary deposits are close to the fluorapatite **[15]**.

b. The wastewater

Before using, the pretreated wastewater undergoes a primary settling for 1 day, which can significantly reduce the pollution load.

Pollution parameters that were analyzed are; turbidity using a turbidimeter WTW according to NF T 90-053 standard, suspended solids with a set of vacuum filtration according to NF T 90-105 the chemical oxygen demand (COD) according to NF T 90-101 and this standard with a reactor VELP Scientifica, biological oxygen demand (BOD) using a WTW Oxytop and NF T 90-103. Table 2 summarizes the results:

Table 2 : Characterization of waste water pretreated

Settings	Raw water
pH	8,10
Conductivity (µs/cm)	3,39
Turbidity (NTU)	312
MES (mg/l)	776
COD (mg/l)	1034
BOD_5 (mg/l)	833

c. Description of the pilot:

The treatment will be made on a glass column with 50 cm of height and 10 cm of diameter, the filter media has a height of 15 cm, it is arranged over 3 cm of gravel, everything is laid on a filter worn by a grid allowing the free flow of the treated water **[16,17]**. The column is made up in its end by a funnel with a hose, which has 1cm of diameter through which the water outlet is recovered. The filter medium is crowned with a layer of 3 cm of gravel and a canvas in order to ensure the dispersion of the feed water on the entire bed and to avoid the formation of a preferential path **[16]** (Fig.1). The clarified water is contained in a tank from which it is withdrawn at a rate of 0.5 l/h.

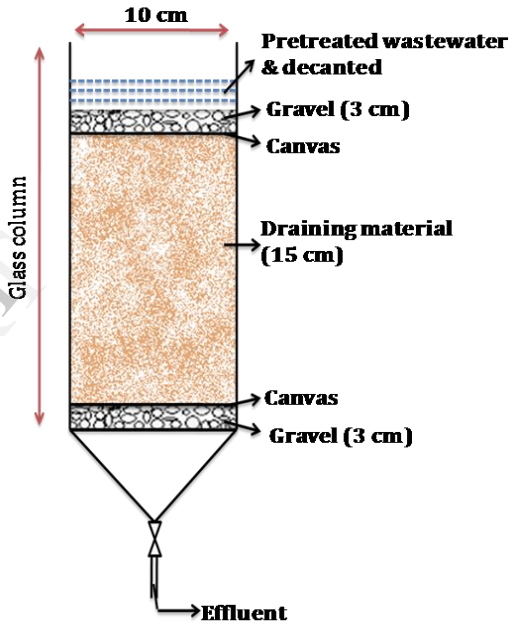

Fig 1 : Drawing of the processing pilot

When the pressure drop, turbidity, or both reach their maximum values, the filter was washed in distilled ensuring continuous water supply. Washing was stopped when the turbidity of the inlet water is equal to that of water at the outlet.

d. Radioactivity measurements

Disk shaped CR-39 (manufactured by Pershore Mouldings Ltd., U.K) and LR-115 type II (manufactured by Kodak Pathé, France and marketed by Dosirad, France) solid state nuclear track detectors (SSNTDs) of radius q=2cm have been separately placed in close contact with wastewater and phosphate samples in hermetically sealed (using a glue) cylindrical plastic containers for one month (30days).

During this exposure α-particles emitted by the nuclei of the ^{238}U and ^{232}Th series inside the considered material samples bombarded the SSNTD films. After the irradiation, the exposed films were etched in two NaOH solutions at optimal conditions of etching, ensuring good sensitivities of the SSNTDs and a good reproducibility of the registered track density rates: 2.5N at 60°C for 120 minutes for the LR-115 II films and 6.25N at 70°C for 7 hours for the CR-39 detectors **[18]**.

After this chemical treatment, the track densities registered on the CR-39 and LR-115 II SSNTDs were determined using an optical microscope. Backgrounds on the CR-39 and LR-115 II SSNTDs were evaluated by placing these films in empty well-closed plastic containers identical to those used for analysing material samples for one month and counting the resulting track densities. This operation was repeated ten times: track densities registered on the CR-39 and LR-115 II detectors were found to be identical within the statistical uncertainties. As the system is well-sealed (there is no escape of radon and thoron) and the exposure time was 30 days, one can assume radioactive secular equilibrium between uranium and each of its decay products and between thorium and each of its daughters.

For our experimental etching conditions, the residual thickness of the LR-115 type II detectors measured by means of a mechanical comparator is 5µm. This thickness defines the lower (E_{min} = 1.6MeV) and upper (E_{max} = 4.7MeV) energy limits for registration of tracks of α-particles in LR-115 type II films. All α-particles emitted by the uranium and thorium series that reach the LR-115 detector at an angle lower than its critical angle of etching θ_c' with a residual energy between 1.6 MeV and 4.7 MeV are registered as bright track-holes. The CR-39 detector is sensitive to all α-particles reaching its surface at an angle smaller than its critical angle of etching θ_c. The critical angles of etching θ_c' and θ_c were calculated by using a method described in detail by Misdaq et al. **[19]**.

The global track density rates (tracks.cm^{-2}.s^{-1}), due to α-particles emitted by the uranium and thorium series inside a material sample, registered on the CR-39 (ρ_G^{CR}) and LR-115 II (ρ_G^{LR}) detectors, after subtracting the corresponding backgrounds, are respectively given by **[18]**:

$$\rho_G^{CR} = \frac{\pi q^2}{2 S_d} C(U)\, d_s \left[A_U \sum_{j=1}^{8} k_j R_j \varepsilon_j^{CR} + A_{Th} \frac{C(Th)}{C(U)} \sum_{j=1}^{7} k_j' R_j' \varepsilon_j^{'CR} \right] \quad (1)$$

and

$$\rho_G^{LR} = \frac{\pi q^2}{2 S_d'} C(U)\, d_s \left[A_U \sum_{j=1}^{8} k_j R_j \varepsilon_j^{LR} + A_{Th} \frac{C(Th)}{C(U)} \sum_{j=1}^{7} k_j' R_j' \varepsilon_j^{'LR} \right] \quad (2)$$

where S_d and S_d' are respectively the surface areas of the CR-39 and LR-115 II films, C(U) (ppm) and C(Th) (ppm) are the uranium (^{238}U) and thorium (^{232}Th) concentrations of the material sample, A_U(Bq/g) = 0.0123 and A_{Th}(Bq/g) = 0.0041 are the specific activities of a sample for a ^{238}U content of 1ppm and a ^{232}Th content of 1ppm, d_S is the density of the material sample (g cm^{-3}), R_j and R_j' are the ranges, in the sample, of an α-particle of index j and initial energy $E_{\alpha j}$ emitted by the nuclei of the ^{238}U and ^{232}Th series, respectively, k_j and k'_j are respectively the branching ratios corresponding to the disintegration of the nuclei of the ^{238}U and ^{232}Th series and ε_j^{CR}, $\varepsilon_j^{'CR}$, ε_j^{LR} and $\varepsilon_j^{'LR}$ are respectively the detection efficiencies of the CR-39 and LR-115 II detectors for the emitted α-particles. The first terms (right of eqs. (1) and (2)) correspond to the number of α-particles emitted by the ^{238}U family (8 α-emitting nuclei), whereas the second terms correspond to the number of α-particles emitted by the ^{232}Th series (7 α-emitting nuclei).

Combining Eqs. (1) and (2), measuring ρ_G^{CR} and ρ_G^{LR} track density rates and calculating ε_j^{CR}, $\varepsilon_j^{'CR}$, ε_j^{LR} and $\varepsilon_j^{'LR}$ by using a "SSNTDEαM" Fortran programme **[18]** one can evaluate the ^{238}U (C(U)) and ^{232}Th (C(Th)) contents inside the considered wastewater and phosphate samples.

3. Results and discussion

^{238}U and ^{232}Th contents have been measured in different phosphate, wastewater, and gravel samples by using equations (1) and (2). Data obtained are shown in Table 3. Data obtained for the ^{238}U and ^{232}Th concentrations in wastewater, phosphate and gravel samples.

Table 3 : Results of radioactivity

Samples	ρ_G^{LR} x10^5 (tr cm^{-2} s^{-1})	ρ_G^{CR} x10^5 (tr cm^{-2} s^{-1})	C (^{238}U) ppm	C (^{232}Th) ppm
Before using in pilot of infiltration				
Untreated wastewater	2.96±0.24	9.11±0.73	0.78±0.06	0.93±0.07
Unused gravel	19.15±1.53	61±5	6.7±0.54	17.90±1.43
Unused phosphate	465±37	1360±108	157±13	11.00±0.97
After using in pilot of infiltration				
Treated water	3.11±0.28	9.55±0.86	0.82±0.07	0.95±0.09
Used gravel	17.36±1.56	55±5	6.3±0.57	17.40±1.57
Used phosphate	449±40	1313±118	152±11	10.50±0.95

According to the results presented in Table 2 it is noted that the concentrations of ^{238}U and ^{232}Th in water relatively increased after passing through the filter of phosphate. This can be explained by the presence of calcite and apatite in phosphate samples **[20]**. We also noted a decrease of the concentration of uranium and thorium in the phosphate and gravel used in the treatment; this could be explained by the transfer of these radionuclides from phosphate to water.

The WHO standards for drinking water set a limit of 0.015 mg/l for uranium, while for thorium which is relatively insoluble in water concentration rarely exceeds 1 µ/l (4mBq/l). As a result, the dose of radioactivity present in the treated water by infiltration percolation on a bed of phosphate is tolerable compared to the standards established by the International Commission on Radiological Protection **[20, 21]**.

4. Conclusion

In this study, we showed the transfer of radioactivity from phosphate rock to the treated water using the system for wastewater treatment by infiltration percolation through a bed of phosphate

However, the water's concentration of uranium (0.82 ± 0.07 ppm), thorium (0.95 ± 0.09 ppm), and those of phosphate (C (^{238}U) = 152 ± 11 ppm and C (^{232}Th) = 10.50 ± 0.95) have shown that the wastewater treated by phosphate is not contaminated by uranium and thorium. Therefore, it does not represent any dangerous action to the environment.

5. Acknowledgments

The authors wish to thank "the Moroccan Association for the Advancement of Science and Technology" (MPST Association) to have participated in the development of this work.

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