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Application of Gamma Irradiation Technique and Aquatic Plants for Wastewater Treatment

Helal Ragab Moussa,[a] Gehad Genidy Mohamed,[b] El Said A. Nouh and [c] Reda Elsayed Abd El Wahed[b]

[a]Radioisotope Department, Nuclear Research Center, Atomic Energy Authority,
[b]Chemistry, Department, Faculty of Science, Cairo University, P.O. Box 12613, Giza, Egypt,
[C]Nuclear Materials Authority, P.O. 530 El Maadi, Cairo, Egypt.

Abstract: With the increase in population in many of the big cities and the growing industrialization in many countries, the pollution load on the environment was increasing, especially heavy metals and pathogenic bacteria, parasites and viruses. Biosorption and removing some heavy metals such as Cd(II), Pb(II) and Cu(II) ions from the aqueous solutions by dried leaves of *Typha domingensis* was investigated, The results showed that the removal of Cd(II), Pb(II) and Cu(II) ions percentages increased as pH, contact time and concentration increased. The results revealed the ability of *Typha domingensis* to remove heavy metals from the aqueous solutions but there was a distinctive variation in biosorpency. Pb(II) ion was the most adsorbed among other metal ions then Cd(II) ion, and the lowest was Cu(II) ion. It was found that the adsorption process of Cd(II), Pb(II) and Cu(II) ions followed second order rate kinetics. The utilization of ionizing gamma irradiation techniques of sewage water offers an efficient, simple and reliable method to produce pathogen free wastewater. This study demonstrated the potential of gamma irradiation to disinfect sewage to increase the water quality in the wastewater by lowering the total heterogeneous bacteria.

Keywords: Biosorption, Heavy metals, Adsorption, Gamma irradiation, *Typha domingensis.*

1. Introduction

Water pollution is considered as major environmental problem worldwide. Contaminations by heavy metal ions are the strongest problem due to high toxicity and they cannot be destroyed by biological degradation. The use of natural materials as adsorbent for heavy metals removal has become a concern in all countries. Many methods were used to remove heavy metal ions such as chemical precipitation, coagulation, adsorption, ion exchange, membrane filtration, solvent extraction and liquid membrane [1-4]. These methods have some disadvantages, as they do not always provide a satisfactory removal rate to meet the pollution control limits. Application of these technologies is so expensive and using of these above mentioned methods may introduce a solid toxic waste in most of cases. Many natural materials were used for waste water treatment such as dried leaves water hyacin [5], *Coconut husk* [6], *Carpobrotus edulis* plant [7], *Eichhornia crassipes* [8], Duckweed [9], Aquatic plant cattail [10], *Ruppia maritima* and *Echinodorus amazonicus* [11], *Neem* leaves powder [12], maize (*Zea mays*) leaf [13], *Fontinalis antipyretica* [14], *platanus orientalis* leaves [15], maple wood sawdust [16], *Tridax procumbens* leaves [17], *Spirodela intermedia*, *Lemna minor* and *Pistia stratiotes* [18], tree leaves [19], dehydrated wheat bran [20], biomaterials chitosan, coffee, green tea, tea, aloe, and Japanese coarse tea [21], saw dust [22], Azolla plant [23], *Canna indica* [24,25], E*ichhornia crassipes* [26], bael tree leaf powder [27] and nile rose plant [28].

Sewage wastewater contains various pathogenic organisms that are capable to induce several human diseases. The disinfection mechanism is explained by two major effects, the direct effect (damage of DNA) and the indirect effect (reaction with free radical). Gamma radiation induces production of free radicals which in turn cause denaturation of cell protoplasm and damage of membranes and cell walls. These processes lead to lysis of organisms [29]. Gamma radiation has been recognized as a new method to eliminate pathogens in sewage sludge [30- 32].

Accordingly, in this study we aimed to investigate the adsorption potential of typha plant for the removal of Cd(II), Pb(II) and Cu(II) ions from wastewater within various experimental conditions. In the same time, this can be considered a way of recycling such environmentally trouble maker materials. Also, we investigate the use of ionizing gamma radiation for waste water treatment. These techniques of treatment to sewage water offer an efficient, simple and reliable method to produce pathogen-free wastewater, low concentrations of contaminants, and removing environmental toxicity.

2. Materials and Methods

Materials

All chemicals were of analytical grade. Copper(II), lead(II) and cadmium(II) standard solution were prepared by stepwise dilution from standard stock solution 1000 mg L^{-1}, (CPA Chem, Bulgaria) in deionized water.

Corresponding Author: Helal Ragab Moussa, *helal_moussa@hotmail.com*

Hydrochloric acid and sodium hydroxide (Merck, Germany) were used to adjust pH. Nutrient agar, Macconkey's Agar, Sabouraud dextrose agar and biochemical media (catalase, urease, Indol, triple sugar) were purchased from (Oxoid, England), oxidase (Oxoid, UK). Gram stain was obtained from (Bio Stec, Egypt).

Preparation of adsorbent material

The adsorbent material was the dry leaf of *typha* (*Typha domingensis).* It was collected from fresh water streams river Nile (Geziret El-Dahab). The plant samples were collected and stored in polyethylene bags and then transported to the laboratory in an icebox within the limited time. Typha leaf was cutted, washed with tap water followed by deionized water, dried on oven at 70 ºC for 48 hours, then the dried materials were ground into powder and preserved in paper bags.

Metal Analysis

Metal ions concentrations in solutions were determined by Atomic Absorption Spectrometer (Thermo Electron Corporation-S series) with deuterium lamp background correction.

Batch method
Effect of pH

The adsorption of Pb(II), Cd(II) and Cu(II) ions under varied pH was carried out at 25 ^{o}C and 100 rpm shaking. 0.25 g of dry typha plant was added to 50 ml of Pb(II), Cd(II) or Cu(II) ions solution (1 mg L^{-1}) at different pH ranging from 2 to 8 and contact time 60 min. The pH values were adjusted using 0.1 N HCl or 0.1 N NaOH. The experiments were conducted until reaching equilibrium and then filtrated. The residual concentration of Pb(II), Cd(II) and Cu(II) ions was determined using atomic absorption spectrometer.

$$\mathbf{q_e = \frac{(Co - Ce)}{w} \times v} \quad \mathbf{(1)}$$

Where q_e (mg g^{-1}) is the uptake of the Pb(II), Cd(II) and Cu(II) ions at equilibrium, C_O (mg L^{-1}) and C_e (mg L^{-1}) are the initial and equilibrium concentration of metal ion, respectively, W (g) the weight of dry plant and V the volume of solution used in liter.

Effect of contact time

The adsorption of Pb(II), Cd(II) and Cu(II) ions as a function of contact time at room temperature was achieved by varying the contact time and keeping the other parameters such as pH, temperature, weight and initial concentration constant. 0.25 g of dry plant typha placed in serial of flasks (8 flask, one for each time interval 5, 10, 15, 30, 45, 60, 90 and 120 minutes) each contain 50 ml of metal ion solution (1 mg L^{-1}) and the content of the flask was shaken at 100 rpm at pH 5, 7 and 6 for Pb(II), Cd(II) and Cu(II) ions, respectively. After reaching the time interval of each flask, the solution was filtrated and then the residual concentration of Pb(II), Cd(II) and Cu(II) ions (C_e) was determined using flame atomic absorption spectrometer.

Effect of concentration

The adsorption of Pb(II), Cd(II) and Cu(II) ions as a function of concentration at room temperature was achieved by varying initial concentration of Pb(II), Cd(II) and Cu(II) ions and keeping the other parameters such as pH, temperature, weight and contact time constant. 0.25 g of dry plant typha was added to 50 ml Pb(II), Cd(II) and Cu(II) ions, placed in serial of flasks (6 flasks, one for each initial concentration of metal ion 0.1, 0.3, 0.5, 1, 2, 5 and 10 mg L^{-1}) and the content of flasks were shaken at 100 rpm at room temperature, pH 5, 7, 6 and contact time 30, 45, 45 min for Pb(II), Cd(II) and Cu(II) ions, respectively. Samples were filtered and then the residual concentration of Pb(II), Cd(II) and Cu(II) ions, (C_e) of each flask was determined.

Bacterial identify of wastewater

Study area

The study area is located in the industrial zone of 10^{th} of Ramadan city in Sharkia governorate, Egypt. Sewage water samples were collected from wastewater treatment plant. The wastewater was exposed to primary and secondary treatments. The wastewater samples were collected from two sites of the plant, the first site is the plant inlet (raw sewage water) and the second site is the plant outlet (treated sewage water). The water samples were collected at five time intervals during 24 hour and mixed to form composite samples (2 L). The wastewater samples were collected in sterilized bottles. After collection, all samples were put on ice and kept at 4 °C until arriving at the lab. For bacteriological assay.

Gamma irradiation

The raw and treated sewage water samples were exposed to gamma radiation. The sewage wastewater samples were exposed to 0.0, 15, 20, 25 KGy. Irradiation was performed through the use of a Canadian Gamma Cell-40 (137Cs) at the National Center for Radiation Research and Technology, Nasr city, Cairo, Egypt. The dose rate was 0.6 Gy/minute.

Characterization of bacterial isolates

Bacterial strains were previously isolated from wastewater samples and identified by routine morphological, microbiological and biochemical methods [33]. Use of blood and Mackonkey agar and biochemical was to identify of bacteria (urease, triple sugar, catalase, indol, oxidase), and sabouraud agar was for fungi. Colonies were removed from these plates and sub-cultured for isolation and identification of bacterial species. It was done by recording macroscopic and microscopic characters. The purified colonies were subjected to Gram staining and characterized using biochemical tests and consulting the pertinent literature and total viable count [34-37].

Total viable count

Samples which obtained from wastewater treatment plant to examine the inactivation of microorganism at various gamma irradiation doses (0.0, 15, 20 and 25 KGy) of the plant inlet and the plant outlet wastewater, total coliform bacteria was counted using pour plate method [38].

3. Results and discussion

Effect of pH

The adsorption of Pb(II), Cd(II) and Cu(II) ions on typha plant as a function of pH was shown in Figure. (1). At room temperature, the uptake capacity increases as pH increase. The maximum uptake (q_e) of Pb(II) ion obtained at a pH range of 5 - 6 and then slight decrease up to pH 6. The maximum uptake (q_e) of Cd(II) ion obtained at a pH range of 7 - 8. The maximum uptake (q_e) of Cu(II) ion obtained at pH range of 6 - 7.

The lower the pH, the more H^+ ions competing with metal ion for adsorption sites, thus reducing their adsorption. On the other hand, the higher the pH, the less the H^+ ions competing with metal ions for adsorption sites, thus increasing their adsorption, which explains the obtained results [39].

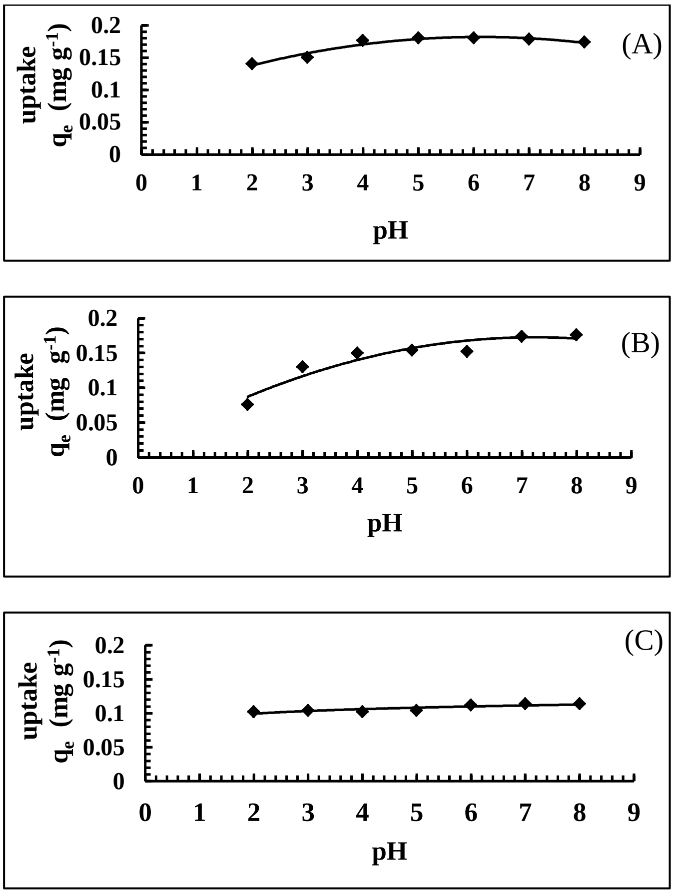

Figure (1). Effect of pH on the uptake of (A) Pb(II), (B) Cd(II) and (C) Cu(II) ions from initial concentration (1 mg L^{-1}) at 25 °C using typha plant.

Effect of contact time

The adsorption of Pb(II), Cd(II) and Cu(II) ions as a function of contact time using initial pH 5, 7 and 6 for Pb(II), Cd(II) and Cu(II) ions, respectively and at room temperature was shown in Figures (2). The maximum Pb(II), Cd(II) and Cu(II) ion adsorption was achieved within 30, 45, 45 min, respectively. Percent removal of Pb(II), Cd(II) and Cu(II) ions increase as contact time increase. This result is important, as equilibrium time is one of the most important parameter for selecting wastewater treatment.

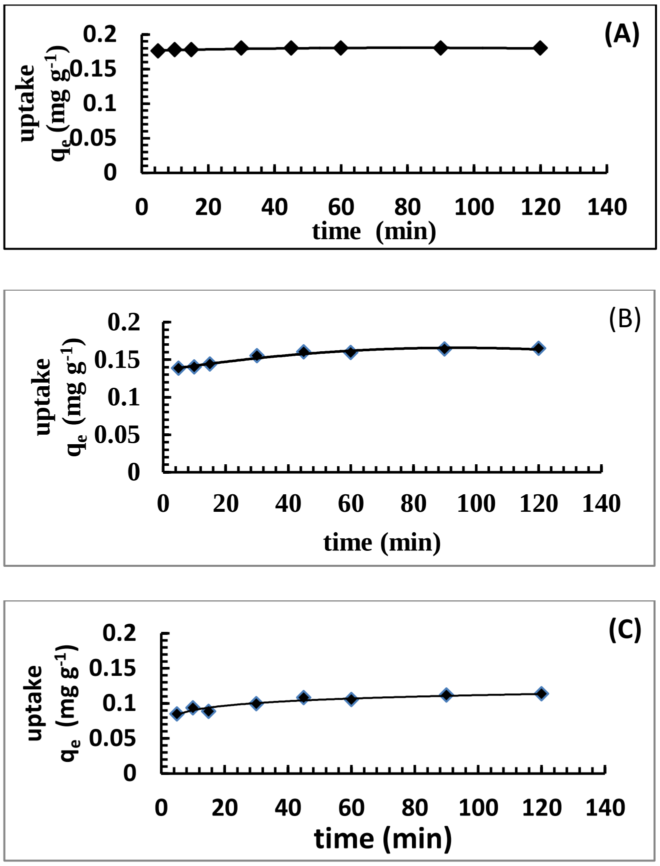

Figure (2). Effect of contact time on uptake of (A) Pb(II), (B) Cd(II) and (C) Cu(II) from initial concentration (1 mg L^{-1}) using typha plant.

Kinetic studies

The adsorption/time data obtained were applied to two kinetic models [40], include pseudo first order and pseudo second order. The pseudo first order expressed as.

$$\log (q_e - q_t) = \log q_e - (k_1/2.303)\, t \qquad (2)$$

Where k_1 is pseudo first order rate constant (min^{-1}), q_e and q_t (mg g^{-1}) refer to amount of metal ion adsorbed at equilibrium and at time t, respectively.

Pseudo second order expressed as.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \qquad (3)$$

Where k_2 (g mg^{-1} min^{-1}) is pseudo second order rate constant of adsorption. The validity of the model was checked by calculating the correction coefficient value of straight lines (R^2) as well as consistence between experimental and calculated value of q_e. The adsorption of Pb(II) processes was found fit well pseudo second order more than pseudo first order model as shown in Figures (3-5). The good fit of kinetic data in Pseudo second order rate expiration show excellent linearity with high correlation coefficient.

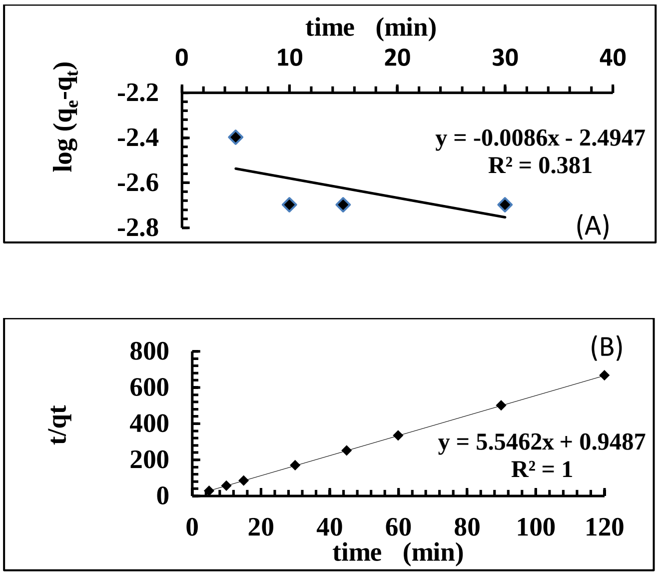

Figure (3). The adsorption of (A) pseudo second order and (B) pseudo second order Pb(II) ion from intial concentration (1 mg L^{-1}), at 25 ^{o}C and rotation speeds 100 rpm.

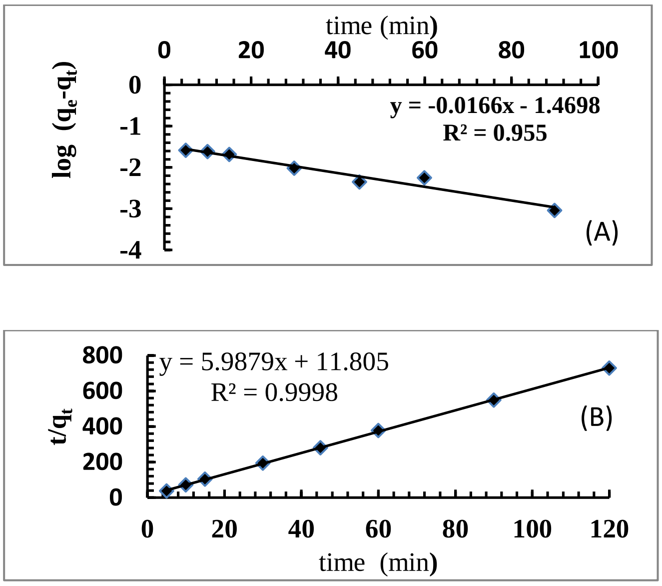

Figure (4). The adsorption of (A) pseudo second order and (B) pseudo second order Cd(II) ion from intial concentration (1 mg L^{-1}), at 25 ^{o}C and rotation speeds 100 rpm.

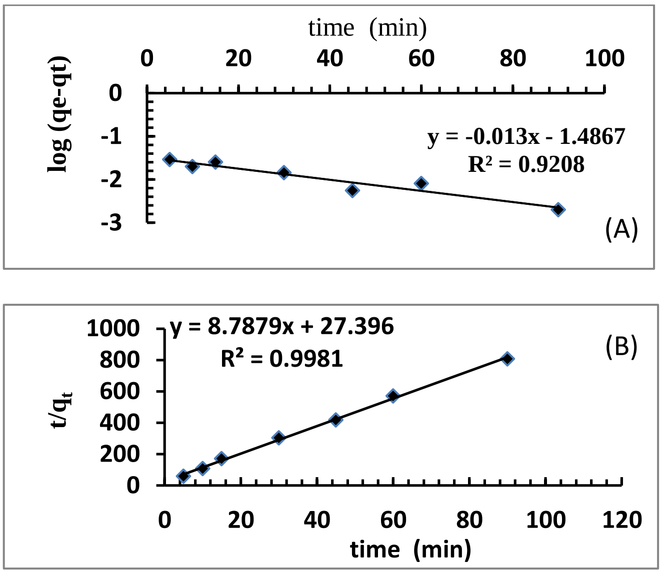

Figure (5). The adsorption of pseudo second order of The adsorption of (A) pseudo second order and (B) pseudo second order Cu(II) ion from intial concentration (1 mg L^{-1}), at 25 ^{o}C and rotation speeds 100 rpm.

Effect of concentration

The adsorption of Pb(II), Cd(II) and Cu(II) ions on typha plant as a function of concentration was shown in Figures (6). The uptake capacity increases as concentration increase.

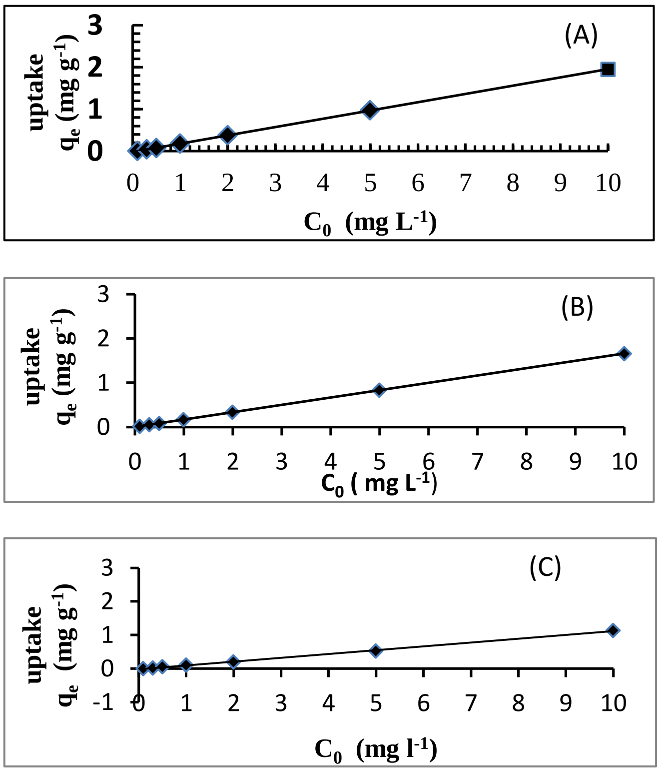

Figure (6). Effect of concentration on uptake of of (A) Pb(II), (B) Cd(II) and (C) Cu(II) ions at 25 ^{o}C, t = 30, 45, 45 min and using initial pH 5, 7 and 6 for Pb(II), Cd(II) and Cu(II) ions, respectavly and rotation speeds 100 rpm, using typha plant.

Characterization of isolated bacteria species

Bacteria species found on waste water included *Staphylococcus aureus, Klebsiella, Pseudomonnas, Escherichia coli* and *Proteus species) and* fungi (*Candida albicans* and *Aspergillus brasiliensis).*

Total viable count

Figure (7) showed the inactivation of coliform (organisms) in the sewage water before treatment samples at various radiation doses. Figure (8) showed the inactivation of coliform (organisms) in the sewage water after treatment of samples at various radiation doses. Figure (9) showed the plot of the logarithmic of colony forming units (CFU)/ml for effluent samples collected from sewage water after treatment chamber versus gamma dose.

The D_{10} value is defined as the dose required to achieve a reduction of 90% of bacteria and was calculated from equation (4).

$$D_{10} = \frac{\text{Radiation dose}}{\log(X_O - X)} \qquad (4)$$

D_{10} value equal (2.8) KGY.

Where X_o is the initial number of organisms, and X is the number of organisms surviving the radiation dose.
Inactivation from 10^7 to 10^3 of viable count of bacteria
= 2.7 X 4.0 (log reduction) = 11.2 KGY.

D_1 for inactivation of viable count of bacteria from 10^7 to 10^1 of viable count of bacteria = 2.7 X 6.0 (log reduction) = 16.8 KGY
It was found that a 16.8 kGy dose is generally suitable for the wastewater inactivation of bacteria.

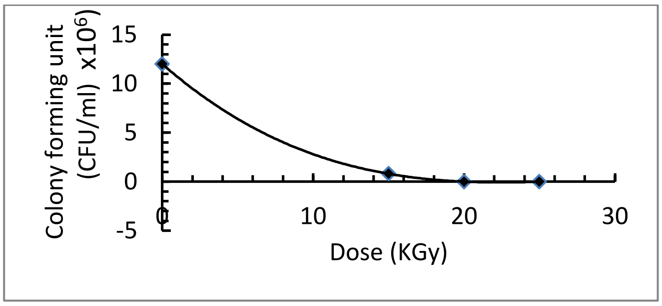

Figure (7). Inactivation of colony forming unit (CFU/ml) from sample of inlet of sewage water by different radiation doses.

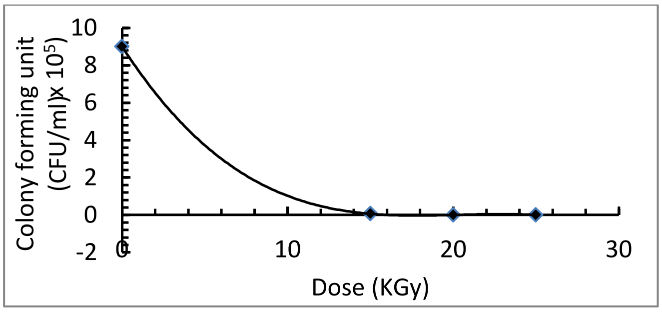

Figure (8). Inactivation of colony forming unit (CFU/ml) from sample of outlet of sewage water by different radiation doses.

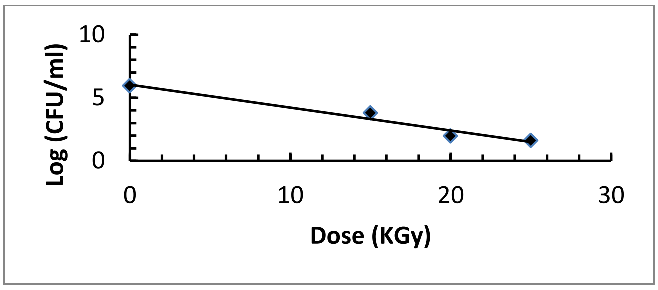

Figure (9). log of colony forming unit (CFU/ml) from sample of outlet of sewage water by different irradiation doses.

4. Conclusion

The presented study showed that *Typha domingensis* offers several advantages including cost effectiveness, high efficiency and minimization of heavy metal ions especially Cd(II), Pb(II) and Cu(II) ions after their adsorption. Biosorption by the dried leaves of Typha domingensis is a technique can be used for removing metal pollutants from water.

The result of batch adsorption clarifies that the percentage of removal metal ions from their solution increase with increase pH, contact time, initial metal ion concentration.

The results of the pH study showed that maximum sorption occurred at pH 5-6. The percentage of Pb(II) adsorbed is slightly lower at higher pH. The maximum uptake (q_e) of Cd(II) ion was obtained at a pH range of 7-8 while the maximum uptake (q_e) of Cu(II) ion was obtained at pH range of 6-7. The results of contact time showed that the maximum adsorption of Pb(II), Cd(II) and Cu(II) ions were 30, 45 and 45 min, respectively.

The adsorption of Pb(II), Cd(II) and Cu(II) ions processes were found fit well pseudo second order more than pseudo first order model, where the good fit of kinetic data in pseudo second order rate expiration showed excellent linearity with high correlation coefficient.

The following bacterial and fungal species were found on wastewater before treatment they included bacteria, (*Staphylococcus aureus, Klebsiella, Pseudomonnas, Escherichia coli* and *Proteus* species*) and* fungi (*Candida albicans* and *Aspergillus brasiliensis*)**.**

The results obtained after treatment with gamma radiation showed the inactivation of coliform (organisms). in the sewage water after treatment samples at various radiation doses 15, 20 and 25 kGy. Radiation treatment technology represented a viable solution to the problem of waste water treatment. It is expected that the spread of use of this technology will provide the impetus for the accelerator industry to produce specially designed for this application. It is hoped that the increase in number of units produced would help the cost of such technology to decline to a level that makes it affordable and economically attractive to many countries.

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Author Profile

Dr. Helal Ragab Moussa
Professor in Plant Physiology and Nuclear Techniques
Radioisotopes Department, Atomic Energy Authority,
Malaeb El-Gamaa St., P.O. 12311, Dokki, Giza, Egypt.