

BIOSORPTION OF AM-241 AND CS-137 BY RADIOACTIVE LIQUID WASTE BY COFFEE HUSK

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ABSTRACT

Radioactive Waste Management Laboratory of Nuclear and Energy Research Institute, IPEN - CNEN/SP, has stored many types of radioactive liquid wastes, including liquid scintillators, mixed wastes from chemical analysis and spent decontamination solutions. These wastes need special attention, because the available treatment processes are often expensive and difficult to manage. Biosorption using biomass of vegetable using agricultural waste has become a very attractive technique because it involves the removal of heavy metals ions by low cost biosorbents. The aim of this study is to evaluate the potential of the coffee husk to remove Am-241 and Cs-137 from radioactive liquid waste. The coffee husk was tested in two forms, treated and untreated. The chemical treatment of the coffee husk was performed with HNO₃ and NaOH diluted solutions. The results showed that the coffee husk did not showed significant differences in behavior and capacity for biosorption for Am-241 and Cs-137 over time. Coffee husk showed low biosorption capacity for Cs-137, removing only $7.2 \pm 1.0\%$ in 4 hours of contact time. For Am-241, the maximum biosorption was $57,5 \pm 0.6\%$ in 1 hours. These results suggest that coffee husk in untreated form can be used in the treatment of radioactive waste liquid containing Am-241.

1. INTRODUCTION

The liquid radioactive wastes at Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP) are mainly generated from nuclear research centers. Several techniques can be employed to treat these wastes, such as precipitation, ion exchange resins and electrochemical processes. However, for removing low radionuclides ion concentrations in large solution volumes, these techniques are not effective for economic reasons [3,4]. Thus, treatment that removes radionuclides from diluted liquid wastes has direct attention to biosorption. It involves the use of biological materials such as microorganisms [11], agricultural wastes [2] and polysaccharide materials [7] to bind the biomass and heavy metal ions by physic-chemical processes: complexation, coordination, chelation, adsorption, ion-exchange and microprecipitation [14].

As natural biosorbent, agricultural residue is getting more attention due to its low cost. Among the biomasses to remove heavy metals in wastewater, the efficiency of coffee husk has already been described [13]. Coffee husk is insoluble in water and it is composed by cellulose (23.08%), hemicelluloses (23.9%), total lignin (28.3%) and ash (0.80%).

The biosorption capacity of the coffee husk can be improved by chemical treatments. Several authors reported a significant increase in the ability of metal biosorption by chemically modified biomass to increase the number of active sites and the presence of new functional groups. [5].

The biosorption of Cs-137 using agricultural residue was investigated only by Mirsha et al [12]. The authors observed that rice hulls removed 17% of Cs after 24 hours of contact time.

For Am 241, the uptake was reported using microbiological biomasses such as *Rhizopus arrhizus* [8], *Candida sp* [13] e *Saccharomyces cerevisiae* [9] with almost 100% of removal from aqueous solution.

Removal behavior of Am-241 and Cs-137 from radioactive liquid wastes was assessed using untreated and treated coffee husks

2. METHODS

2.1. Preparation and Modification of Coffee Husks

The coffee husks used was provided by the Group Sara Lee. Coffee husk was washed with distilled water, dried in oven at 80 ° C for 24 hours, sterilized by UV radiation, crushed and sieved. The fraction between 0.297 mm and 0.500 mm were used at the biosorption experiments. Coffee husks were chemically treated form according to the procedure described by Kumar et al [6].

For the chemical treatment process, 25g of coffee husks were suspended in 1 liter of 0.5 M HNO₃ solution for 3 h at 25 ° C, and stirred by mechanical agitation at 240 rpm. The biomass was filtered off and washed with deionized water 3 times and suspended in a solution of 0.5 M NaOH stirred by mechanical agitation at 240 rpm for 3 hours. After the basic treatment, the biomass was filtered through a Buchner funnel, washed with deionized water until neutral pH and dried at 40 ° C.

2.2. Batch Biosorption Experiments

For biosorption experiments 0.2 g of coffee husks were added in plastic bottles with 10 mL of radioactive liquid waste with initial activity of 11 Bq/mL for Cs-137 and 129 Bq/mL for Am-241 at pH= 4. The vials were shaken on an orbital shaker (100 rpm) at room temperature. After the contact time, the solutions were filtered through filter paper and filtrate was analyzed to assay the residual Am-241 and Cs-137 in solution using a gamma spectrometer, GX2518- Canberra, The assays were performed in different contact times (30 min, 1 h, 2 h and 4 h) and americium ion concentrations. All experiments were run in triplicate.

3. RESULTS

The removals of Am-241 and Cs-137 by untreated and treated coffee husks from radioactive liquid wastes are shown in Fig.1 and 2.

The results showed that both coffee husks forms did not showed significant differences in behavior and capacity for biosorption for Am-241. The maximum removal was circa 57,5 ± 0.6% after 1 hour of contact time (Fig.1 and Tab. 1).

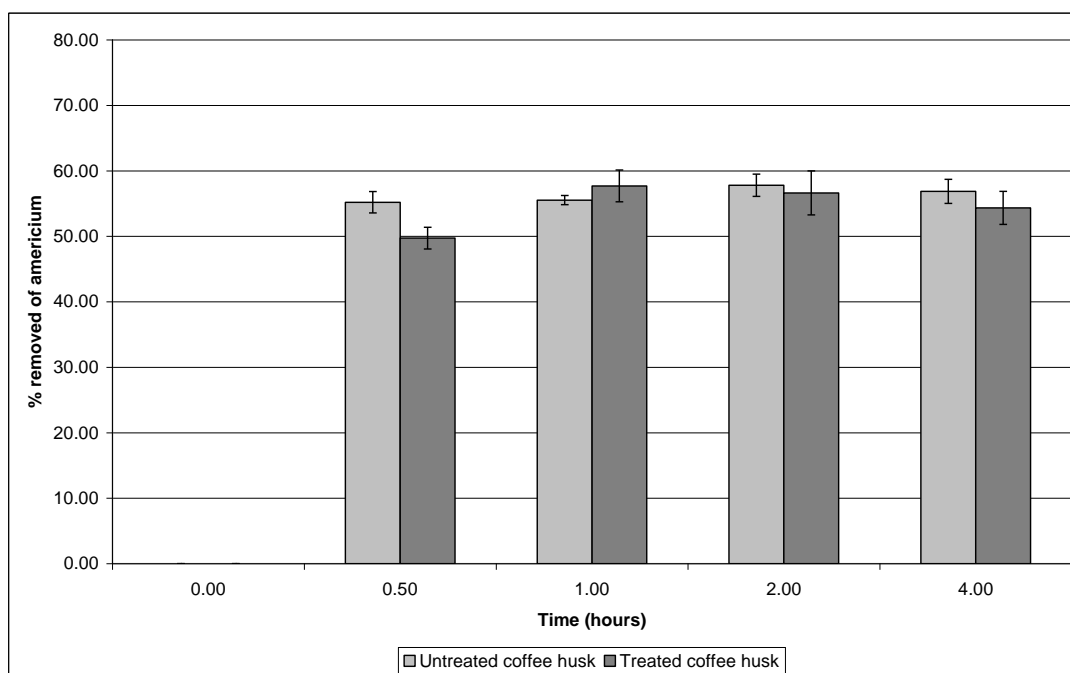


Figure 1: Biosorption of Am-241 with initial activity of 124 Bq/ml at pH 4 by coffee husks

Table 1: Percentage of Americium removed by untreated and treated coffee husks

Time (hours)	Fraction removed (%±sd)	
	Untreated	Treated
0.50	55.20±1.65	49.73±1.63
1.00	55.53±2.43	57.69±0.69
2.00	57.59±3.36	56.62±1.70
4.00	56.86±2.51	54.34±1.82

On the other hand, cesium biosorption was affected by the chemical treatment of the coffee husks (Fig.2). The maximum Cs-137 removal for treated coffee husks reached $7.24 \pm 1.00\%$ after four hour of contact time, while for untreated coffee husks, the maximum removal was $12.95 \pm 2.89\%$ in two hour of contact time (Tab.2). After that time, desorption process is observed. It is noteworthy that in both coffee husks forms, the capacity to uptake cesium from radioactive liquid waste was very low comparing to Am-241.

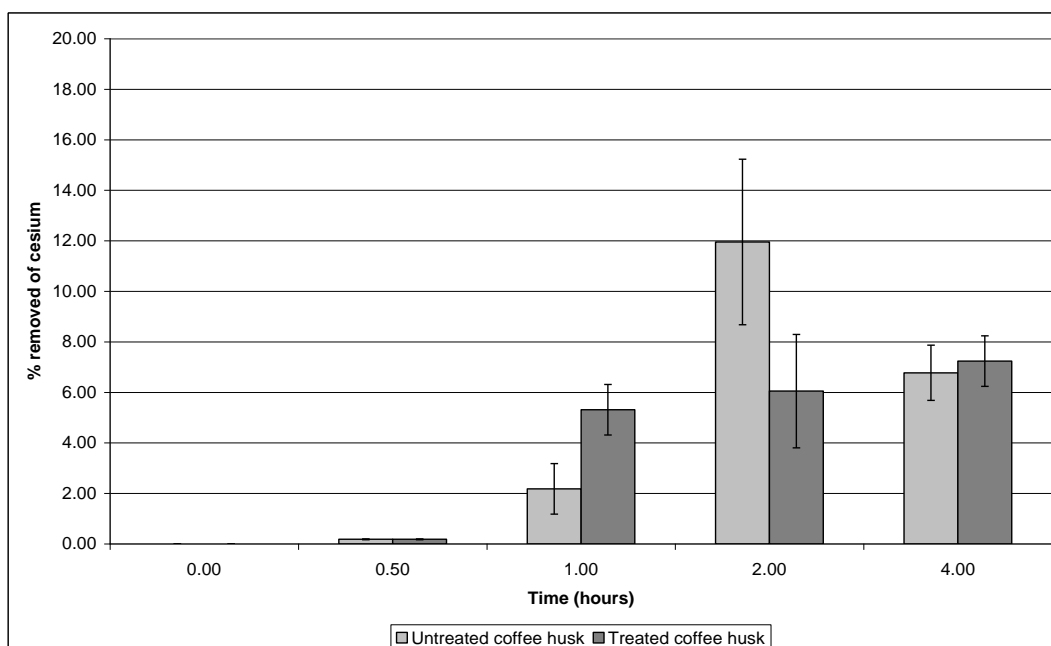


Figure 2: Biosorption of Cs-137 with initial activity of 11 Bq/ml at pH 4 by coffee husks

Table 1: Percentage of Cs-137 removed by untreated and treated coffee husks

Time (hours)	Fraction removed (%±sd)	
	Untreated	Treated
0.50	0.00	0.00
1.00	2.18±1.00	5.31±1.00
2.00	11.95±2.89	6.05±2.24
4.00	6.77±1.09	7.24±1.00

4. CONCLUSIONS

Because there was no significant difference between the results from untreated and treated coffee husks to remove Am-241, the natural form can be used to remove this radionuclide from liquid radioactive waste stored at the Radioactive Waste Management Laboratory IPEN-CNEN/SP. To improve the uptake of Cs-137, other parameters such as, pH, contact time and concentration need to be optimized. The desorption process can be only evaluate after infra red spectroscopy experiments of the biomass in order to analyze the functional groups involved in the bind biomass-metal mechanism.

REFERENCES

1. Bhatnagar, A.; Vilar, V.J.P.; Botelho, M.S.; Boaventura, R.A.R. Coconut-based biosorbents for water treatment – A review of the recent literature. *Advances in Colloid and Interface Science*, **v.160**, p.1-15, 2010. 10
2. Blázquez, G., Martín-Lara, M. A., Tenorio, G., & Calero, M. Batch biosorption of lead(II) from aqueous solutions by olive tree pruning waste: Equilibrium, kinetics and thermodynamic study. *Chemical Engineering Journal*, **v.168(1)**, pp.170-17, 2011.

3. Hiromoto, G.; Dellamano, J.C.; Marumo, J.T.; Endo, L.S.; Vicente, R.; Hirayama, T. "Introdução à gerência de rejeitos radioativos", Instituto de Pesquisas Energéticas e Nucleares, Departamento de Rejeitos Radioativos, São Paulo, 1999.
4. IAEA -International Atomic Energy Agency. Combined methods for liquid radioactive waste treatment. **TECDOC-1336**, Vienna, 2003.
5. Krishnani, K. K., Meng, X., & Boddu, V. M. Fixation of heavy metals onto lignocellulosic sorbent prepared from paddy straw. *Water Environment Research*, **v.80(11)**, pp. 2165-2174, 2008.
6. Kumar, U., & Bandyopadhyay, M.. Fixed bed column study for cd(II) removal from wastewater using treated rice husk. *Journal of Hazardous Materials*, **v. 129(1-3)**, pp.253-259 2006.
7. Li, Y., Liu, F., Xia, B., Du, Q., Zhang, P., Wang, D. Removal of copper from aqueous solution by carbon nanotube/calcium alginate composites. *Journal of Hazardous Materials*, **v.177(1-3)**, pp. 876-880, 2010.
8. Liao, J., Yang, Y., Luo, S., Liu, N., Jin, J., Zhang, T.,. Biosorption of americium-241 by immobilized rhizopus arrhizus. *Applied Radiation and Isotopes*, **v. 60(1)**, pp. 1-5, 2004.
9. Liu, N., Luo, S., Yang, Y., Zhang, T., Jin, J., & Liao, J. Biosorption of americium-241 by saccharomyces cerevisiae. *Journal of Radioanalytical and Nuclear Chemistry*, **v. 252(1)**, p.187-191, 2002.
10. Luo S., Liu N., Yang Y., Zhang, T., Jin, J., Liao J. Biosorption of americium-241 by Candida sp.. *Radiochimica Acta*, **v. 91(6)**, pp. 315-318, 2006. 113
11. Ma, Y., Lin, J., Zhang, C., Ren, Y., & Lin, J. Cd(II) and As (III) bioaccumulation by recombinant escherichia coli expressing oligomeric human metallothioneins. *Journal of Hazardous Materials*, **v.185(2-3)**, pp.1605-1608, 2011.
12. Mishra S P, Prasad S K, Dubey R S, Mishra M, Tiwari D, Lee S M., Biosorptive behaviour of rice hulls for Cs-134 from aqueous solutions: a radiotracer study. *Appl Radiat Isot.*, **v.65(3)**, pp.280-6, 2007.
13. Oliveira; W.E, Franca; A.S., Oliveira; L.S., Rocha; S.D., Untreated coffee husks as biosorbents for the removal of heavy metals from aqueous solutions, *J. Hazard. Mater.* **v.152**, p. 1073–1081, 2008.
14. Wan Ngah, W. S., & Hanafiah, M. A. K. M. Removal of heavy metal ions from wastewater by chemically modified plant wastes as adsorbents: A review. *Bioresour Technol.*, **v.99(10)**, pp. 3935-3948, 2008.