

THE POTENTIAL OF COCONUT FIBERS IN RAW AND TREATED FORMS TO REMOVE ^{241}Am FROM AQUEOUS SOLUTIONS

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ABSTRACT

In the Radioactive Waste Management (GRR) at the Nuclear and Energy Research Institute (IPEN/CNEN-SP) vegetal biomasses has been studied as adsorbent to remove radioisotopes from radioactive liquid wastes as part of the radioactive waste treatment program. In this work coconuts fiber was evaluate as biosorbent to remove ^{241}Am from aqueous solutions and many different parameters were studied such as particle size (35 and 80 mesh) and contact time (between 5 and 60 minutes). In order to evaluate if the treated biomass could remove more ^{241}Am the experiments were also performed using raw biomass and treated with H_2O_2 in basic conditions. When the experiment was carried out using raw coconuts fiber with 80 mesh, neutral conditions after 5 minutes of contact time 99% of the radionuclide was removed from the aqueous solution. This result shows the potential of this biomass to remove ^{241}Am from radioactive liquid wastes.

1. INTRODUCTION

Nowadays radioisotopes are used widely in industries, reactors and research activities. However, radioactive wastes are produced in the process in the handling these radioisotopes. Radioactive wastes are defined as waste that contains or is contaminated with radionuclides at concentrations or activities greater than clearance levels as established by the regulatory body [1], thought no further use is foreseen. If these radioactive wastes are released without a proper treatment or safe control, that can result in impacts to humans and the environment. For this reason, a radioactive waste management is necessary [2].

There are many kinds of radioactive wastes, they are classified according to its physical, chemical and radiological aspects. This classification serves to aim suitable treatment, as well as the safety aspects [2]. To highlighted, are the liquid radioactive wastes, its detection is relatively simple, but the remove of these metals in aqueous solution is still a challenge [3].

Between the technologies studied for the removal of metals, the biosorption is one the most effective on the last years. Biosorption is the process of immobilization of the metals presents in aqueous solution for biosorbents [4]. This technique has the advantage to remove the metals in low concentration [5] and doesn't have the production of sludge in the end of the process.

Studies showed that the alginate (biopolymer) and *Saccharomyces cerevisiae* (fungus) can remove almost of all ^{241}Am present in aqueous solution [6, 7]. Others materials used as biosorbent are the agricultural wastes such as rice, straw, straw, bark, japanese green tea, wool and coconut fiber [8]. These wastes are promising for its low cost and metal-binding capacities [9, 10].

It has been reported that the use of coconut fiber has showed good removal of heavy metals as Cu [11], Ni, Zi, Pb, Th and U [12]. The coconut fiber could be used in raw form or chemically modified to improve its capacity to remove heavy metals [11].

This work aims to removal of ^{241}Am present in aqueous solution using the coconut fibers in raw and treated forms. The particle size, pH and contact time were analyzed to demonstrate the adsorption efficiency.

2. EXPERIMENTAL PROCEDURE

2.1- Pre-treatment and chemical treatment of the biomass

The raw coconut fibers (*Cocosnucifera L.*) were washed with distilled water to remove impurities. After washing, they were dried at 80° C for 24 h and sterilized with germicidal lamp for 2h and separated using a particle size sieves (Granutest). The fraction used was 0.500 mm (35 mesh) and 0.177 mm (80 mesh).

For the chemical treatment process, 10 g of fibers were suspended in 200 ml solution containing 1.5 g of hydrogen peroxide (50%) (Vetec) and 0.1 g of the sodium hydroxide (Dinamica) at room temperature. The temperature was slowly raised to 60°C and oxidation continued for more 2 hours. The material was filtered, washed thoroughly with hot water, rinsed in cold water and then dried at 70°C for 24 h [11].

2.2 – Biosorption process

The biosorption process was performed in batch with under constant agitation on ashaker (Timer) at room temperature using different contact times. After the contact, the solution was filtered through filter paper (Macherey-Nagel). The ^{241}Am remaining in solution was analyzed by using Gamma Spectrometer - Canberra, Model GX2518. All experiments were performed in triplicate.

2.3 – Materials

The stock solution of Americium at concentration about 3007.08 Bq/ml was prepared by dissolving with deionized water to a final concentration of about 200 Bq/ml. The pH was adjusted with dilute solution of NaOH (Dinamica) and HNO_3 (Alphatec) using paper indicador (Macherey-Nagel). The coconut (West Garden) was bought at local trade.

3. RESULTS AND DISCUSSION

The following parameters were studied: contact time, pH influence, particle size of biomass and dosage effect.

3.1 – Effect of particle size

These experiments were carried out using solution with about 200 Bq/ml of ^{241}Am , pH 3, about 0.1g of biomass and 5ml of solution, in contact time of 30 minutes as shown in Table 1:

Table 1: Percent removed of ^{241}Am under influence of particle size of biomass

Nº Exp	Particle Size	Raw Biomass	Treated Biomass
1	35 mesh	84	92
2	80 mesh	50	95

According to PINO (2005), larger particles with spherical shapes, in general, have greater adsorption due to an increased mass transfer of small particles.

3.2 – Effect of dosage

In order to check the best dosage between quantity of biomass and solution, studies were carried out using different amount of biomass with 35 mesh for 5 ml of solution with about 200 Bq/ml of ^{241}Am . The results are shown in table 2:

Table 2: Influence of dosage of biomass to remove ^{241}Am

^{241}Am Uptake (%)			
Nº Exp	Biomass (g)	Raw Biomass	Treated Biomass
1	0.1	84	92
2	0.3	91	92
3	0.5	94	95

The studies showed that the removal of ^{241}Am ions increases with increased mass biosorbent. It is also verified that the amount remaining of solution using by treated biomass is the minor compared to raw biomass.

3.3 – Effect of pH

The influence of pH was carried out using by the best dosage quantity (biomass with 35 mesh) and with solution about 200 Bq/ml of ^{241}Am per 30 minutes of contact time is show in table 3:

Table 3: Influence of pH on the removal of ^{241}Am

^{241}Am Uptake (%)			
Nº Exp	pH	Raw Biomass	Treated Biomass
1	3	92	95
2	5	93	96
3	6	92	98

The pH is one of the most important variables in the removal of heavy metals by biomass due to metal ion capacity to be in a precipitated or complex form in basic condition [3].

Studies of the precipitated or complex form of the radionuclides in solution carried out by ROMANCHUK et.al. (2013), show that, in pH 5 there is a greater amount of Am^{+3} free in solution and the beginning of the formation of AmOH^{2+} . When the pH of the solution is 6, these two complex form are in same amount. As the pH is greater, the amount of Am^{+3} decreases and the amount of AmOH^{2+} increase.

The biomass oxidation converts the cellulosic hydroxyl groups to the carboxyl groups, thus creating a weak cationic ion-exchanger [11], favoring, therefore, the bond of $-\text{COO}^- \text{Am}^{+3}$. The active centers are likely to be positively charged at very low pH values because of high proton concentration, hence hinders the adsorption of metal cations on these binding sites [13, 14, 15].

3.4 – Effect of contact time

The studies of contact times were carried out using 0.5g of raw biomass (figure 1) and treated biomass (figure 2) with 35 mesh and 5 ml of solution about 200 Bq/ml of ^{241}Am with pH 5.

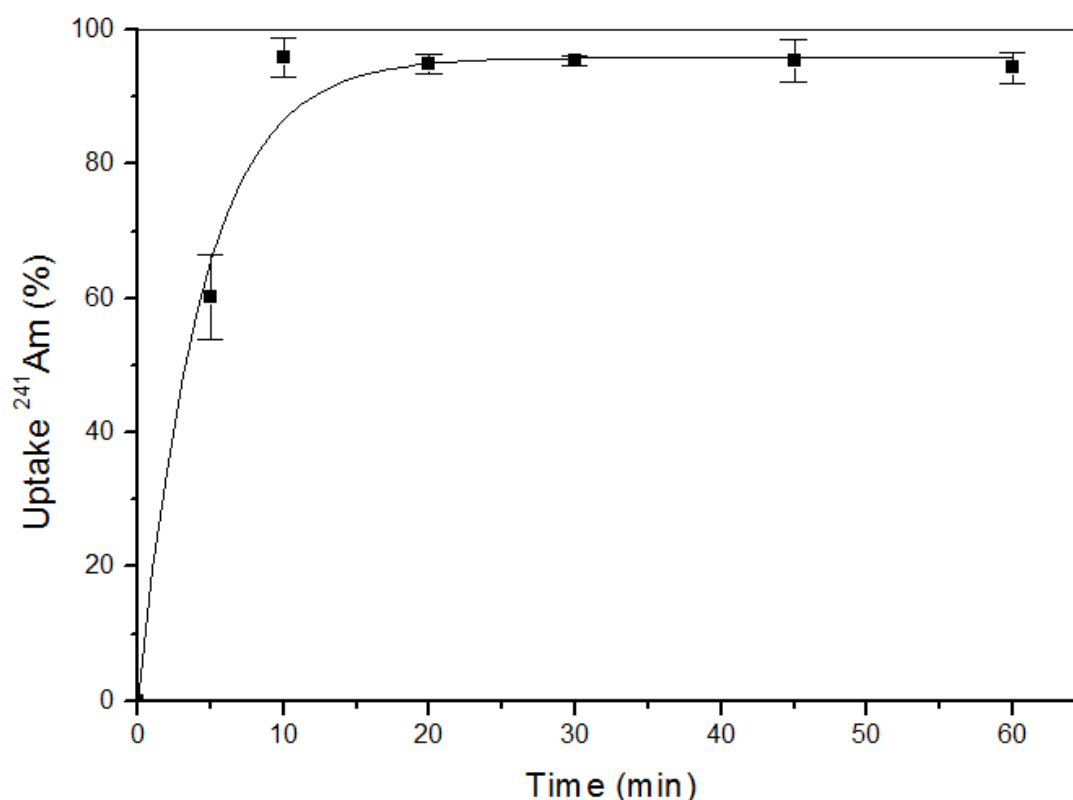


Figure 1 - Effect of contact time; ■ ^{241}Am uptake on raw coconut fiber.

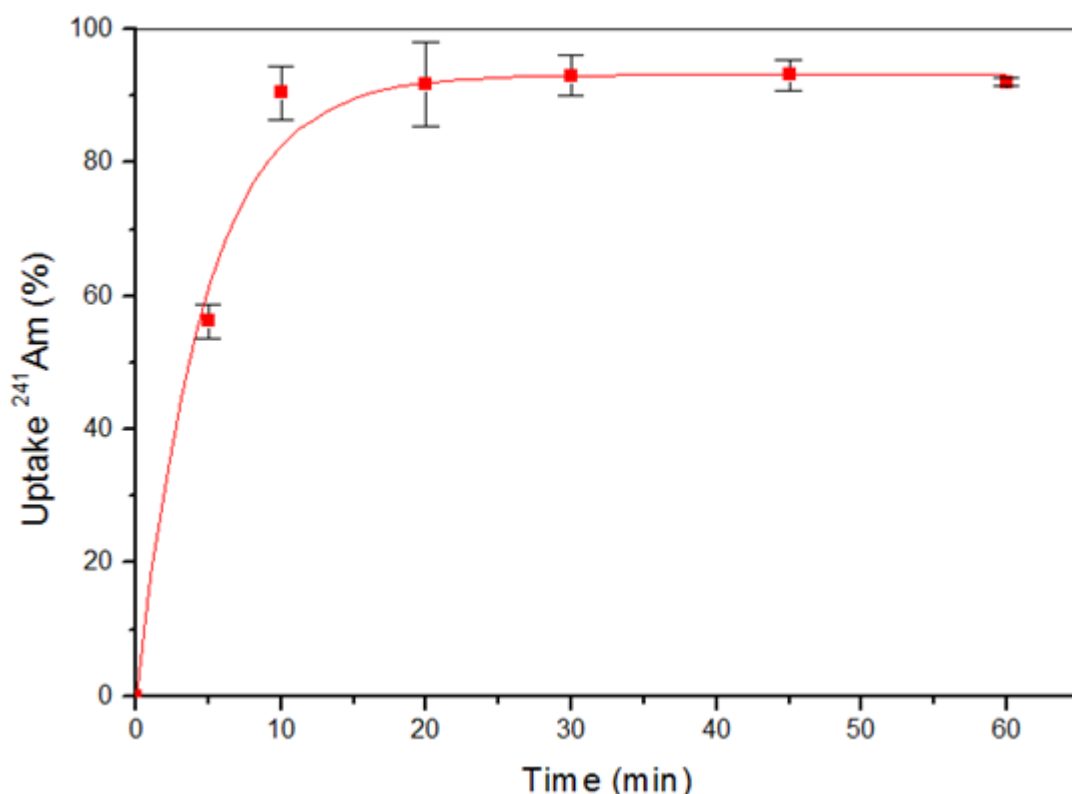


Figure 2 - Effect of contact time; ■ ^{241}Am uptake on treated coconut fiber.

The experiments show that the uptake of ^{241}Am in 10 minutes is about 93% for both biomass (raw and treated), after this time, there is a small increase in removal capacity. In 30 minutes there is an apparent equilibrium.

4. CONCLUSIONS

Comparing the results obtained in the studies performed for removing ^{241}Am using by coconut fiber, is possible concluded that the best conditions are: using treated biomass with particle size of 35 mesh at a dosage of 0.5 g of coconut fiber to 5 ml solution of ^{241}Am about 200 Bq/ml at pH 6 for 30 minutes of contact time. The percentage of ^{241}Am removed under these conditions is about 98%.

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REFERENCES

1. INTERNATIONAL ATOMIC ENERGY AGENCY, *Radioactive Waste Management Glossary*, Vienna (2003).

2. 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, Brasil (1999).
3. A. Y. ROMANCHEK, A. S. ALESAREV, S. N. KALMYKOV, D. V. KOSYNKIN & J. M. TOUR, “Graphene Oxide for Effective Radionuclide Removal,” *Phys. Chem. Chem. Phys.*, **15**, pp. 2321 (2013).
4. S. SCHIEWER & M. H. WONG, “Ionic Strength Effects in Biosorption of Metals by Marine Algae,” *Chemosphere*, **41**, pp. 271-282 (2000).
5. N. AZOUAOU, Z. SADAOU, A. DJAAFRI & H. MOKADDEM, “Adsorption of Cadmium from Aqueous Solution onto Untreated Coffee Grounds: Equilibrium, kinetics and Thermodynamics,” *Journal of Hazardous Materials*, **184**, pp. 126-134 (2010).
6. T. R. BORBA, J. T. MARUMO, M. M. GOES, R. V. P. FERREIRA & S. K. SAKATA, *Biosorption of Americium by Alginate Beads*, International Nuclear Atomic Conference – INAC, Rio de Janeiro, Brasil (2009).
7. R. V. P. FERREIRA, J. T. MARUMO, M. H. BELLINI, A. J. POTIENS JÚNIOR, A. S. TAKARA, M. M. GOES, T. R. BORBA, C. M. NASCIMENTO & S. K. SAKATA, *Use of Saccharomyces Cerevisiae in Radioactive Waste Treatment*, International Nuclear Atomic Conference – INAC, São Paulo, Brasil (2007).
8. M. MINAMISAWA, H. MINAMISAWA, S. YOSHIDA & N. TAKAI (2004) “Adsorption Behaviour of Heavy Metals on Biomaterials,” *J. Agric. Food Chem.*, **58**, pp. 5606-5611 (2004).
9. G. WANG, J. LIU, X. WANG, Z. XIE & N. DENG, “Adsorption of Uranium (VI) from Aqueous Solution onto Cross-Linked Chitosan,” *Journal of Hazardous Materials*, **168**, pp. 1053-1058 (2009).
10. T. A. KURNIAWAN, G. Y. S. CHAN, W. H. LO & S. BABEL “Comparisons of Low-Cost Adsorbents for Treating Wastewaters Laden with Heavy Metals”, *Science of the Total Environment*, **366**, pp. 409-426 (2006).
11. S. R. SHUKLA, V. G. GAIKAR, ROSHAN S. PAI & UMESH S. SURYAVANSHI, “Batch and Column Adsorption of Cu(II) on Unmodified and Oxidized Coir”, *Separation Science and Technology*, **44:1**, pp. 40-62 (2009).
12. R. A. MONTEIRO, M. YAMAURA. *Avaliação do Potencial de Adsorção de U, Th, Pb, Zn e Ni pelas Fibras de Coco*. Tese de mestrado. Instituto de Pesquisas Energéticas e Nucleares – IPEN. São Paulo, Brasil (2009).
13. G. A. H. PINO, *Biossorção de Metais Pesados Utilizando Pó da Casca de Coco Verde (Cocos nucifera)*, Dissertação de mestrado. PUC do Rio de Janeiro, Brasil (2005).

14. T. AKAR, S TUNALI, "Biosorption Characteristics of *Aspergillus Flavus* Biomass for Removal of Pb(II) and Cu(II) Ions from Aqueous Solutions", *Bioresource Technol.* **97**, pp. 1780-1787(2006).
15. T. KAPPOR, T. VIRARAGHAVAN, D. R. CULLIOMORE, "Removal of Heavy Metals Using the Fungus *Aspergillus Niger*". *Bioresource Technol.* **70**, pp. 95-104 (1999).