Adsorption of Radium 226 by Biological Origin Adsorbents

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Summary

Selected samples of waste microbial biomass used in industrial fermentation processes and wastewater biological treatment plants have been studied for their radium biosorption ability from aqueous solutions. Equilibrium biosorption isotherms have been used to quantify the radium uptake capacity of the various types of biomass which were also compared to two types of activated carbon. Solution pH affected the observed uptake significantly. In general, the biomass types that showed appreciable sorption capacity exhibited maximum uptake between pH 7 and 10. The uptake was reduced considerably at pH 4 and little or no uptake was observed at pH 2. Radium biosorptive uptake capacities of the order of 4.5×10^4 nCi/g, at pH 7 and at an equilibrium radium concentration of 1000 pCi/L, were determined for a mixed culture, while the biomass of *Penicillium chrysogenum* adsorbed 5×10^4 nCi/g radium under the same conditions. The highest uptake value for a sample of F-400 granular activated carbon was 3600 nCi/g at pH 7 and 1000 pCi/L radium concentration. The biosorptive radium uptake of microbial biomass is compared to literature values for other types of adsorbents. The most effective biomass types studied exhibited radium removals in excess of 99% of the radium in solution.

INTRODUCTION

Canada has developed an extensive technological base covering the entire uranium nuclear fuel cycle: mining of uranium ore, preparation of nuclear fuel, operation of nuclear power plants, and storage of spent fuel. Recent (1980) electric generation data indicate that nuclear power generated electricity accounts for approximately 10% of the total electricity generated in Canada and 32% of Ontario's total electricity output.[1]

Climatic conditions make Canadian uranium mines unique in North America since they require the disposal of large volumes of wastewater, along with the solids. Flows of 3.5 to 7.2 million gallons per day have been recorded for one tailings area in Elliot Lake.[2] Wastewater flow consists of process wastewater (milling), mine water, and overflow and seepage from active and inactive tailings areas. Current wastewater treatment consists of lime addition to an effluent pH 7-9. This precipitates most contaminants except ra-

dium-226. Because effluents often contain radium-226 activities two orders of magnitude above the federal standard of 10 pCi/L, some form of radium-226 removal is required.[3] In the current treatment method, radium-226 is coprecipitated as a barium–radium sulfate sludge by addition of $BaCl_2$ to the sulfate-rich effluent. The resulting precipitate settles very slowly in unlined settling ponds, often with residence times of several weeks. Effluents from these settling ponds often result in substantial releases of suspended radium-226 to surface water courses.[3] The recent addition of such physicochemical treatment processes as coagulation and flocculation, have produced, on a pilot scale, effluents consistently meeting the total radium-226 level of 10 pCi/L but not the dissolved level of 3 pCi/L.[4] This improvement followed the suggestion of the Radioactivity Sub-Group of the Federal Government Mining Task Force (1974) for the development of physicochemical treatment systems as an alternative to settling ponds.[5] There is considerable concern about the long-term stability of the barium–radium sulfate sludge since redissolution of the radium will take place if fresh water contacts the sludge.[6,7]

One of the physicochemical processes that has been used very effectively for the sequestering of low levels of contaminants from aqueous solutions is adsorption. Under proper design conditions and following the identification of the appropriate adsorbent, adsorption can be very effective and specific. Vast quantities of drinking water and wastewater have been treated by adsorption to remove trace quantities of toxic pollutants. Furthermore, adsorption installations are compact, simple, easy to operate, and the method for their proper design and control has been developed.[8]

Uranium mine and mill tailings areas overflow consists of a high pH, high in sulfates, low in nitrogen wastewater, containing trace levels of radium-226 $(10^{-9}$ g/L), and low levels of heavy metals.[9] Adsorptive treatment of such a wastewater appears attractive provided radium-specific adsorbents with high radium uptake capacities become available.

A radium adsorption system could replace the present $BaCl_2$ treatment of the tailings wastewater. Such an adsorption system would intake tailings pond effluent, which would pass through a system of adsorption beds where radium would be removed down to the desired effluent concentration. Following saturation of the adsorbent (breakthrough of radium), the bed would be decommissioned temporarily in order to go through a radium recovery cycle. Products of the regeneration cycle would be the regenerated adsorbent and a concentrated solution containing the previously adsorbed radium. Thus, radium can be extracted from a very large volume of wastewater and concentrated in a small and manageable volume of high level waste that can be disposed of appropriately. Adsorptive treatment of seepage flow can also be easily performed in abandoned uranium mill tailings areas.

Microbial biomass has been shown to selectively retain heavy metal ions from an aqueous environment. This ability is exhibited in dead microbial cells as well as in live ones. Previous work has shown the affinity of microbial bio-

mass for uranium and thorium in solution.[10-12] This study represents the first systematic investigation of the uptake of radium by inactive microbial biomass.

Experimental data on radium biosorption, from this study, have indicated that radium can be absorbed not only on conventional organic and inorganic adsorbents but on biological origin adsorbents as well. A brief listing of radium adsorbing capacities of various adsorbents follows (with radium uptake capacity given in pCi/g): natural Zeolite[13] (2800); manganese Zeolite[13] (2100); Zirconium salts[13] (2750); Bio-Rex ion exchange resin[10] (2900); activated carbon (present work) (3500); biomass A (present work) (40,000); biomass B (present work) (75,000). It is evident from this listing that biological origin adsorbents have emerged as a novel class of adsorbents of superior radium uptake capacity compared to conventional adsorbents. It is therefore reasonable to propose that these adsorbents may form the basis for a new technology for radium sequestering.

EXPERIMENTAL

Samples of waste microbial biomass used in industrial fermentations and wastewater treatment processes have been either collected and processed, or produced by fermentation under the same conditions as in industry. The samples were requested from industries in Canada and the United States. Eight different types of biomass were finally available and are listed in Table I. Activated carbons, granular Filtrasorb 400, and powdered Darco S-51 were also tested for comparison.

Radium-226 solutions were prepared from a standard solution of $RaCl_2$ in dilute HCl. Adjustment of pH was effected using HCl (BDH) and NaOH (Fisher) reagent solutions.

Initial solution radium-226 concentrations ranged from 50 to 1000 pCi/mL. Initial solution pH levels were set to 2, 4, 7, and 10. There are potential experimental difficulties when dealing with radium solutions. In neutral and basic solutions, uptake of radium by glass surfaces occurs. When using solu-

TABLE I
Eight Different Types of Biomass Studied

1) *Aspergillus niger*
2) *Penicillium chrysogenum*
3) *Pseudomonas fluorescens*
4) *Streptomyces niveus*
5) *Rhizopus arrhizus*
6) Municipal-wastewater-activated return sludge A
7) Municipal-wastewater-activated return sludge B
8) Industrial-wastewater-activated sludge (Refinery)
9) Filtrasorb 400 granular activated carbon (Calgon)
10) Darco S-51 powdered activated carbon (Atlas Chemicals)

tions of such low radium concentrations, this uptake can be both consider-able and erratic. In an effort to minimize radium uptake by the contact flask wall, flasks made of polyethylene, a material more hydrophobic than glass, were used. All contacts were performed in polyethylene, 500-mL Erlenmeyer flasks with neoprene stoppers. The flasks were agitated on an orbital shaker. The contact solution volume was 100 mL. The standard procedure used for determining activated carbon adsorption isotherms was implemented.[10]

The liquid-to-solid ratio in the contact flasks was varied by using different weights of dry biomass ranging from 10 to 250 mg. The biomass was precon-ditioned by contacting it with water of the appropriate pH and continually adjusting the pH to the desired level until no further significant change in pH occurred. The biomass was then filtered and dried. Biomass preconditioning ensured solution pH stability during contact.

Separation of the sorbents from solution was accomplished by vacuum filtration through 0.45-μm Sartorius (Millipore) cellulose acetate–nitrate membrane filters. Each filter was washed with deionized water before use. The first 50-mL portion of filtrate was used for pH measurement; the second 50-mL portion was used for radium-226 determination. Duplicate contacts, with no sorbent present, were performed for each pH level to determine the extent of radium uptake by the contact flasks.

The contacts were performed at 23°C. The contact time was 24 h for the biomass and 5 days for the activated carbon.[14] The solutions were analysed for radium-226 before and after contact. The radium-226 analyses were per-formed by filling standard 25-mL jars to capacity with sample solution. The jars were then wax sealed to prevent escape of radon gas and stored for 30 days to establish secular equilibrium.

A series of standards was prepared from a radium-226 stock solution (NBS). The 609.4-keV photopeak of bismuth-214 was monitored. At secular equilib-rium, the ratio of bismuth-214 activities in the standard and each sample is identical to the ratio of radium-226 activities.

In general, the size of the containers was such as to provide a counting rate that permitted measurements with ± 5% precision over a 500-to-4000-s count-ing period. The limit of detection for a 4000-s counting period was 1 pCi/mL.

Gamma-ray spectra of all samples and standards were recorded using an Ortec Coaxial Ge(Li) detector shielded by a lead enclosure with a full width at half-maximum of 2.2 keV at the 1.3332-MeV photopeak of cobalt-60, a peak-to-Compton ratio of 30:1, and an efficiency of 10%. The detector was used in conjunction with a Canberra series-30 1024-channel pulse-height analyzer with data output to a teletype.[15]

RESULTS

Loading (uptake) values (q, in nCi/g) were calculated by taking the differ-ence in radium-226 content of the precontact and postcontact solution di-vided by the weight of dry sorbent.

Plots of q versus residual equilibrium concentration of radium-226 remaining in the solution after contact (C_{eq}, in nCi/L), allow a quantitative comparison of the radium uptake capacity of the examined adsorbents. Trend lines on the plots, facilitate the visualization of data behaviour.

Contact Flask Wall Effect

Table II summarizes some representative values of the change in the contact solution radium concentration due to adsorption of radium by the flask walls. Radium has been documented to exhibit a strong adsorption behavior, which, coupled with the low radium solution concentrations used, resulted in deviations from the starting radium solution concentration during the biosorption equilibrium studies. These changes were quantified through the use of two contact flasks containing the standard volume of radium contact solution with no biomass (blanks). The average radium solution concentration of the two blanks at the end of the contact time was used as the initial radium solution concentration for the rest of the contact flasks. The blank contact tests revealed that the use of polyethylene rather than glass flasks reduced, but did not eliminate, the problem of radium uptake by the contact vessel walls. This resulted in some scatter in data points. The contact flask wall effect is considered further in the Discussion section.

Solution pH Effect

For the biomass types that adsorb radium, in general, no significant uptake occurs until the pH is raised to 4. A maximum uptake is observed between pH 7 and 10. This behavior is illustrated by the linearized adsorption isotherms presented in Figures 1-6. Some of the materials examined (e.g. *Pseudomonas fluorescens*), exhibited very low radium uptake under the examined experimental conditions and have not been presented graphically. Their low radium loading resulted in wider scattering of data. The experimental results determined in the present work clearly indicate that the pH range from 7 to 10 is the optimum working pH range for radium biosorption.

TABLE II
Flask Wall Effect on Initial Radium Concentration
Percent Change of Initial Concentration

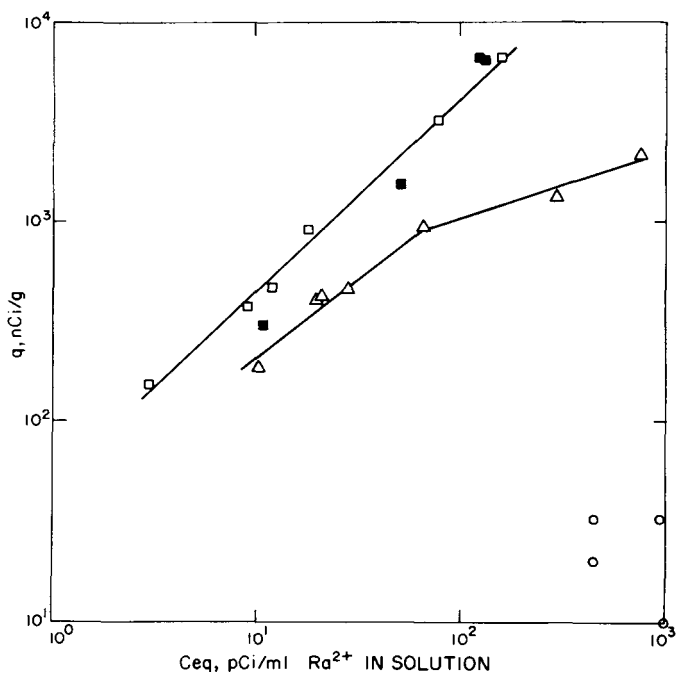| Solution pH	Initial solution radium concentration (pCi/mL)		
	50	500	1000
2	+24	+2	−2
4	−6	−5	−5
7	−44	−14	−13
10	−84	−35	−21

Fig. 1.   Linearized radium-226 adsorption isotherms by inactive biomass of *Penicillium chry-sogenum:* (○) pH 2, (△) pH 4, (□) pH 7, and (■) pH 10.

Differences in uptake between pH 7 and 10 were small and in many cases insignificant. Table III summarizes the observed radium uptake, by all the adsorbents examined, at pH 7 and at three common equilibrium solution radium concentrations.

Initial Radium Concentration Effect

Three different levels of initial radium concentration were examined, namely 50, 500, and 1000 pCi/mL. It is clearly indicated that the observed radium uptake by the biomass samples is a strong function of radium solution concentration. This behavior is typical of physicochemical adsorption systems.

All determined radium equilibrium adsorption isotherms could be linearized on a log–log scale similarly to the case of uranium and thorium adsorption by inactive microbial biomass that has been reported earlier.[10] Furthermore, in certain cases, radium adsorption isotherms presented break points such as those observed in the case of adsorption of organics by activated carbon. Such break points are usually indicative of a change in the equilibrium behavior of the adsorption system.
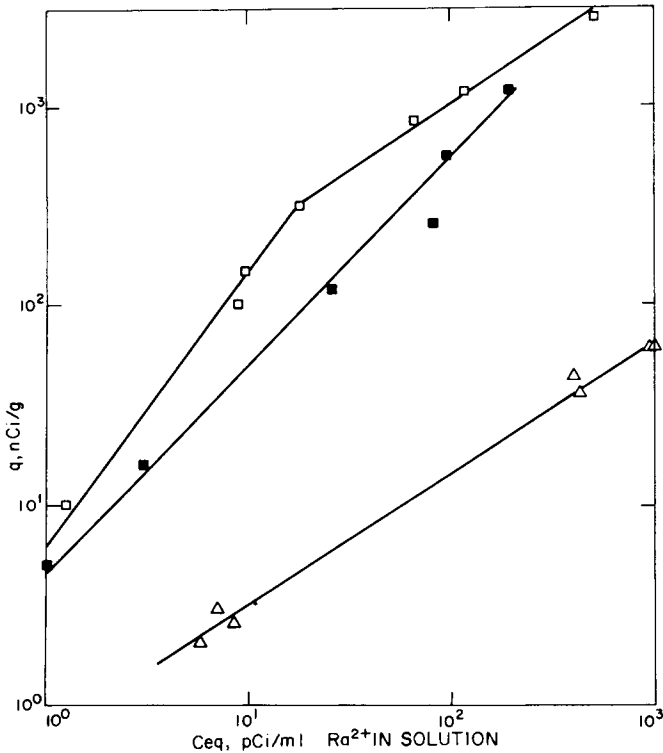
Fig. 2. Linearized radium-226 adsorption isotherms by inactive biomass of *Rhizopus arrhizus:* (△) pH 4, (□) pH 7, and (■) pH 10.

DISCUSSION

Adsorbent Characteristics

Positive radium uptake capacity was exhibited by all adsorbing media examined. However, considerable variation in their radium adsorption capacity was observed (Table III). The data presented thus far, suggest that individual characteristics of each adsorbent affect significantly the corresponding radium adsorption capacity. *Rhizopus arrhizus,* for example, has been demonstrated to be a better adsorbent than both the waste-activated sludge and *Penicillium chrysogenum* for uranium and thorium.[10] The reverse, however, holds for radium adsorption (Figs. 1, 2, and 5). Similarly, some difference in radium adsorptive capacity was exhibited by the two activated carbons tested. Granular Filtrasorb-400 (F-400) (Fig. 7) appeared more effective at higher solution pH values than Darco S-51, although S-51 was in powder form, thus presenting a higher "external" surface area to the radium solution. The higher specific surface area of F-400 (1200 m^2/g vs. 700 m^2/g) and the associated
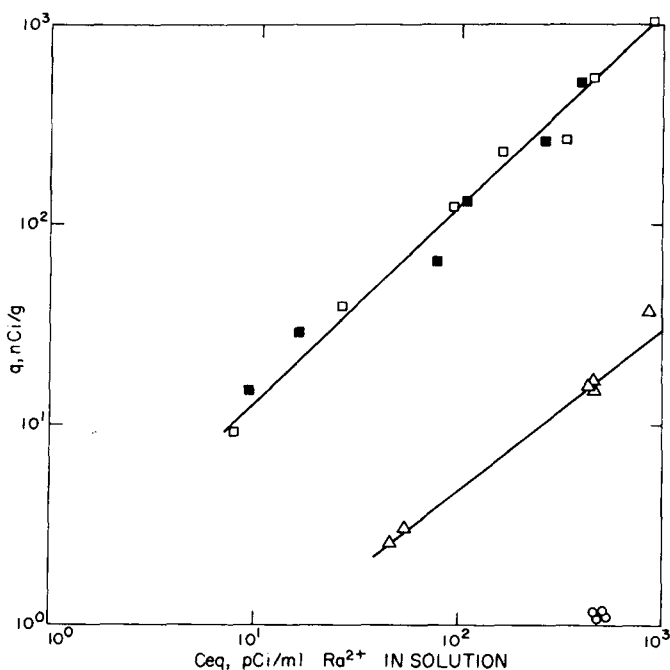
Fig. 3.   Linearized radium-226 adsorption isotherms by inactive biomass of *Aspergillus niger*. Symbols are the same as in Fig. 1.

larger mean pore diameter (35 Å vs. 30 Å) could be considered responsible for the observed difference on the basis of the recently advanced adsorption mechanism hypothesis.[16]

The best two biological origin adsorbents are representative samples of two entirely different microbial biomass types. *P. chrysogenum* is a pure culture (fungal) while municipal wastewater activated sludge biomass is a mixed culture with considerable proportions of bacterial species.[17] Two different samples of municipal activated sludge were collected, processed, and tested for their radium biosorption capacity: one from a small treatment plant in Quebec, used for previous uranium biosorption experiments (A) and another from the City of Hamilton wastewater treatment plant (B). Although differences in the speciation of the two samples should be expected, both samples presented similar radium uptake at pH 7 and 1000 pCi/L radium equilibrium concentration. The behavior of the two activated sludge biomass samples was not very different within the examined concentration and pH range (Figs. 5 and 6), with both types exhibiting uptakes of the same order. This similarity could be justified by considering that the dominant microorganism types present in municipal wastewater treatment plant sludges are usually common, and that these common components are responsible for the observed, generally high, radium uptake capacity.[17] Further support for the above hypothesis is provided by the lower radium uptake capacity exhibited
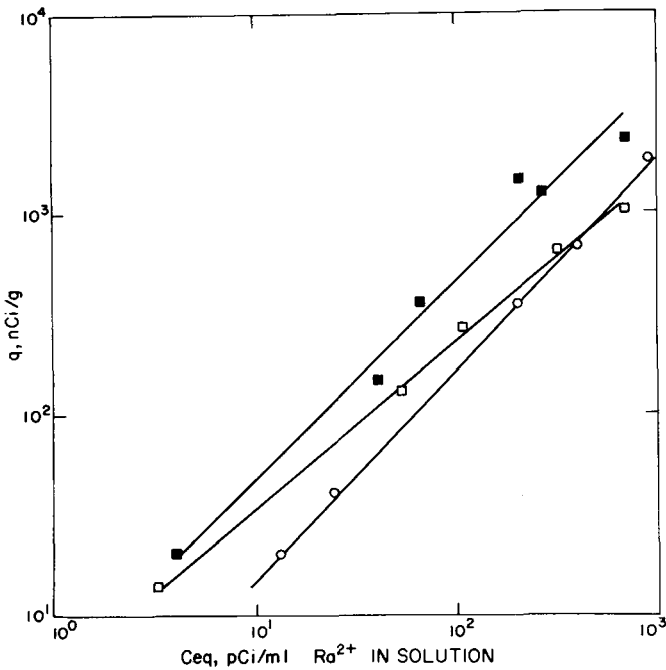
Fig. 4. Linearized radium-226 adsorption isotherms by inactive biomass of *Streptomyces ni-veus:* (○) pH 2, (□) pH 7, and (■) pH 10.

by a mixed culture originating from a refinery wastewater treatment plant (industrial sludge). Evidently the different composition of the industrial wastewater resulted in different speciation of the microbial population of the industrial activated sludge, the components of which, as the experimental data indicate, possess radium uptake capacity lower than the components of the municipal sludge (Table III).

The validity of such a mechanism in radium biosorption by waste activated sludges is significant as it indicates that waste sludge from municipal waste-water treatment plants, presently landfilled or incinerated, can be considered as an inexpensive and efficient radium adsorbent. This adsorbent could be used to develop technical radium adsorbents of desired technical characteristics or even used in the available form to polish effluents contamined with radium. The minimal cost of this material, the availability, and the associated high adsorption capacity [concentration factors (solid phase concentration)/(liquid phase concentration) of 10^5-10^6] make it attractive.

Solution pH Effect

A clear trend was indicated by the experimental results. Radium uptake capacity of all the materials that were tested increased as the solution pH in-
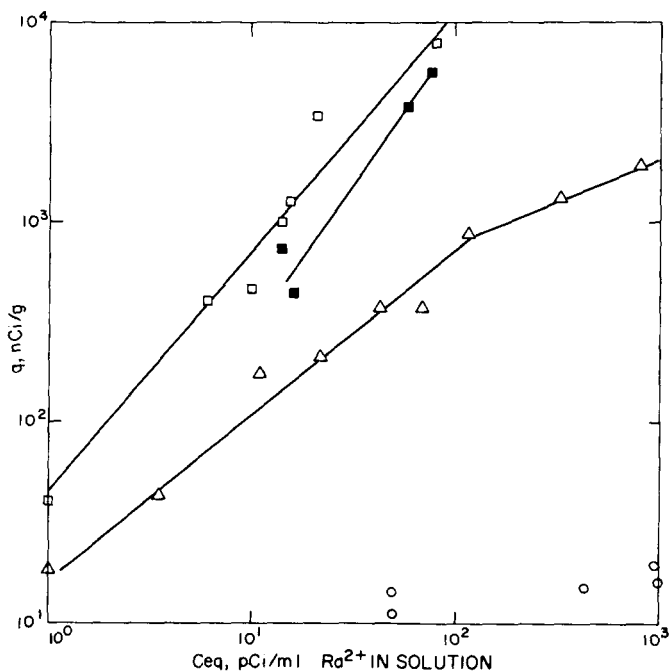
Fig. 5. Linearized radium-226 adsorption isotherms by municipal-wastewater-activated re-
turn sludge A. Symbols are the same as in Fig. 1.

creased towards the neutral and alkaline range. This observation is consis-
tent with the observed increased radium sorption by the contact flask walls as
the solution pH increased. It is suggested that the adsorptivity of radium in
solution increases in the neutral and alkaline pH range. Such an increase in
adsorptivity could be associated with the simultaneous decrease in solubility
observed as the solution pH increases, regardless of the low radium solution
concentrations. Radium, being the heaviest alkaline earth metal, shows the
least tendency to hydrolyze and has the most soluble hydroxide of the ele-
ments in that group of the periodic table.[18] As a result, unlike uranium or
thorium, radium hydrolysis should not be expected to significantly affect the
observed biosorptive behavior. Furthermore, significant differences were not
observed between the radium adsorptive capacity exhibited at pH 7 and pH
10 by the materials tested. Consequently, the neutral pH range, an easier and
environmentally safer pH range, can be considered optimum for radium
removal by biosorption, a fact that facilitates the engineering applications of
the process. The elucidation of the fundamental processes (mechanism) in-
volved in radium sequestering through biosorption will make possible a more
refined understanding of the solution pH effect on the observed radium up-
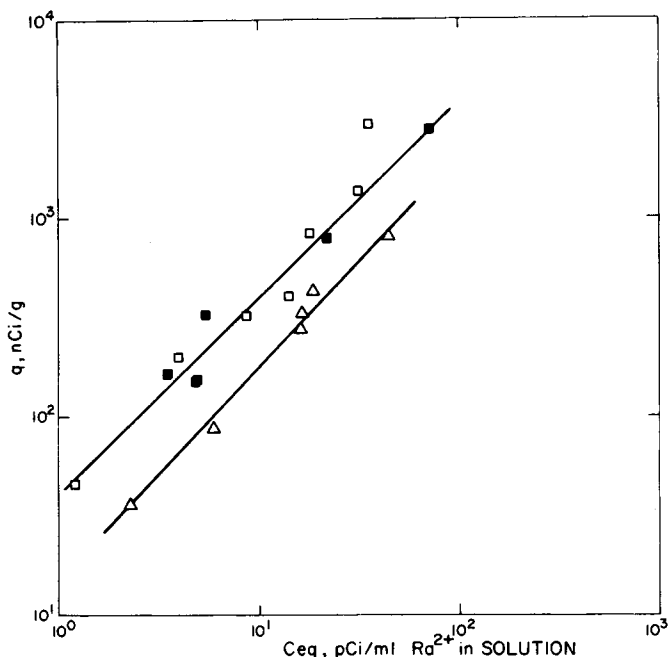take capacity.

Fig. 6. Linearized radium-226 adsorption isotherms by municipal-wastewater-activated return sludge B. Symbols are the same as in Fig. 2.

Initial Radium Concentration Effect

Initial radium concentration had no discernible effect on the determined radium equilibrium biosorption isotherms. Starting with different conditions of initial radium concentration and biomass (adsorbent) dosage the same equilibrium point could be attained. In addition, it can be concluded that the equilibrium points determined are independent of the samples used in each individual experiment as experimental results could be duplicated within the precision of the employed experimental method. Similar conclusions have been reached for the adsorption of both uranium and thorium on biological origin adsorbents from aqueous solutions. [10-12]

Contact Flask Wall Effect

The system—radium solution and contact flask—has to be treated as an ordinary reversible adsorption system. Radium uptake by the flask walls was shown to be a function of the radium solution concentration and pH, as presented in Table II. At the end of each experiment, the contact flask walls were equilibrated with radium, as dictated by the radium solution concentration and pH. Incomplete cleaning of the flask walls between experiments could result in either further leaching of additional radium from the flask
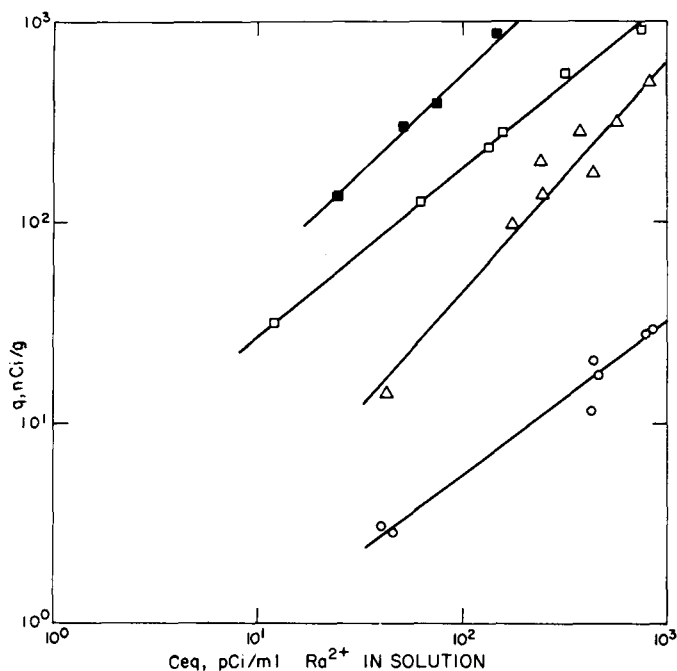
Fig. 7.   Linearized radium-226 adsorption isotherms by granular-activated carbon Filtrasorb 400. Symbols are the same as in Fig. 1.

walls into the radium contact solution, or sorption of radium from the contact solution by the flask walls during the next experiment. Consequently, the hydrophobicity of the flask walls becomes a major factor that may determine the significance of the obtained experimental results. Flasks made of polyethylene, with a critical surface tension of 31 mN/m were used rather than glass (ca. 80 mN/m). However, the experience of the present work has

TABLE III
Radium Uptake Capacities (nCi/g) at pH 7

Radium equilibrium concentration (pCi/mL)	*P. fluorescens*	*A. niger*	*S. niveus*	Industrial sludge	*R. arrhizus*	Return sludge B	*P. chrysogenum*	Return sludge A	S-51	F-400
							Adsorbent type			
10	—	12	32	40	140	390	450	700	(8)[a]	26
50	—	60	130	230	650	1900	2100	5000	(26)[a]	100
300	20	340	580	1700	2000	(11,000)[a]	(12,000)[a]	(40,000)[a]	120	460

[a] Values in parenthesis obtained from linear extrapolation.

clearly indicated that materials of even lower critical surface tension are necessary to ensure better experimental precision. Work presently in progress utilizes flasks made of teflon with a critical surface tension of 18.5 mN/m. In addition, a more rigorous cleaning procedure has been developed, tested, and adopted for the contact flasks so as to lessen the effects of reversible radium adsorption by the flask walls during biosorption experiments. This procedure requires the use of both a complexing agent (ethylenediaminetet-raacetic acid) and HCl solutions. The need for the complicated and cumbersome experimental contact technique that has been developed, can be better appreciated if one considers the very low levels of radium used in this work and the considerable effect the reversible adsorption of radium by the flask walls may exert on the significance of the experimental results.

Comparison to Adsorption Data in Literature

Most of the experimental data reported in the literature refer to inorganic adsorbents such as those summarized in the Introduction. The work of Arnold and Crouse is comprehensive and indicates that most of the inorganic adsorbents tested exhibit, at 1000 pCi/L, radium loadings on the order of 3000 pCi/g.[13] The activated carbon sample tested did not exhibit appreciable radium adsorption capacity. The characteristics of this activated carbon (such as specific surface area or average pore diameter) were not reported by Arnold and Crouse. It has been indicated in the present work that these characteristics may significantly affect the radium adsorption capacity of an activated carbon. It is interesting to note that, as in the present work, lower radium loadings in the acidic solution pH range were also reported.

Manganese-oxide–dioxide-impregnated acrylic fibers were used to extract radium from sea water by Moore and Reid.[19] They experienced low loadings of approximately 4.3 pCi/g of fiber at 1 pCi/L solution radium concentration.

The use of cationic inorganic ion exchangers (zeolites) for radium removal from contaminated drinking water has been reported by Brinck et al.[20] However, their data do not allow the calculation of the equilibrium radium uptake capacity of the ion exchanger used.

In an internal publication of the Czechoslovak Atomic Energy Commission, evidence is presented by Stamberg, Jikek, and Stamberg that the biomass of *Penicillium chrysogenum* can adsorb radium from aqueous solutions to a final loading of 10^3-10^4 pCi/L of wet biomass.[21] Evidence is also presented for the accumulation of radium by a number of bacterial organisms present in radium contaminated natural waters. Although these results cannot be directly compared to the present work, as the related experimental conditions and detailed data have not been published, they serve the purpose of confirming the ability of microbial biomass to sequester radium from dilute aqueous solutions.

In a recent publication by Durham and Joshi, the activated sludge from two Ontario municipal wastewater treatment plants (Hamilton and Dundas) were tested for radionuclide content.[22] Radium loadings of 1024 and 634

pCi/kg, respectively, were determined for the two sludges while the sewage treatment plant effluent was reported to be free of radium. The radium present in the sludge can be considered as the result of radium biosorption from the wastewater inflow. These results confirm the ability of municipal wastewater sludge to adsorb radium from dilute aqueous solutions. The reported uptake values are low because the sludge is at equilibrium with the very low radium concentration of raw sewage (not reported). Complete radium uptake isotherms for the same sludge were determined in the course of the present work and are presented as Figure 6.

CONCLUSIONS

The experimental results of the present work led to the following conclusions. Adsorption of radium by biological origin adsorbents is feasible and in fact such adsorbents may present higher radium loading values than a variety of conventional organic or inorganic adsorbents. As anticipated for a physicochemical adsorption system, observed radium adsorption capacity is a function of radium solution equilibrium concentration and pH. Optimum radium adsorption pH is in the range 7–10. Within the above pH range the change in radium adsorption capacity can be small or insignificant depending on the adsorbent. Radium adsorption at low (pH 2) solution pH is very limited. The biomass of *Penicillium chrysogenum* can be considered as an effective radium adsorbent. Waste activated sludge from municipal wastewater treatment plants can be as good as or better radium adsorbent than *Penicillium chrysogenum*.

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