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Removal of Toxic Metals by Biosorption onto Nonliving Sewage Sludge

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ABSTRACT

Toxic metals, such as zinc, nickel, and cadmium, can be removed from dilute aqueous solutions by sorption onto nonliving sewage sludge applied as finely dispersed biosorbent particles after sterilization and drying. A comparison between the suggested method and precipitation, the most common method used for metals removal, was conducted in parallel. The main parameters examined in single component systems include initial metal concentration, temperature, and inhibition of the removal process by the existence of soluble constituents. Adsorption isotherms were employed to describe the metals uptake; the Langmuir types were found to fit the experimental data better than the Freundlich ones. Moreover, the desorption of metals from metal-loaded biomass was investigated. The possible selective separation of metals from binary and ternary mixtures by biosorption, which could lead to the recovery and recycling of the removed metals, was also examined by introducing an illustrative selectivity factor.

INTRODUCTION

The development of low-cost biological technologies for the bioremediation of industrial effluents is currently of great interest and importance. The sequestering of metal ions by solid materials of biological origin is known under the general term "biosorption." Previous investigations

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were mainly focused on the influence of metal ions to living microorganisms, emphasizing the toxic effects and the subsequent inhibition of the activated sludge process during biological wastewater treatment processes (1–5). Since the initial findings that living activated sludge can remove metals from wastewaters, great progress in research has been realized on the uptake and immobilization of metals by waste biomass, produced during many biotechnological applications, such as *Penicillium chrysogenum* and *Rhizopus arrhizus* (6), and *Streptomyces clavuligerus* and *Streptomyces griseus* (7, 8). In these cases, flotation of metal-laden biomass was applied as the separation process downstream.

Predominant mechanisms by which living microorganisms can facilitate the abstraction of soluble metal ions from aqueous solutions include volatilization, extracellular precipitation, complexing and subsequent accumulation, passive sorption at binding sites on the envelopes of cells, and intracellular accumulation (metabolically mediated uptake) (9). The occurrence of these mechanisms depends on the metals and the types of microorganisms. The composition of the medium may also have a direct effect on both passive adsorption and metabolic uptake. Metal ions in aqueous solution may also be precipitated as insoluble sulfides, produced in-situ, when certain industrial effluents are depleted of oxygen; in these anaerobic environments, sulfate-reducing bacteria are widely distributed. Extracellular biopolymers usually present in activated sludge processes include mainly polysaccharides, but proteins and nucleic acids can also be found and may play an important role in metal–microorganism interactions.

Metal uptake by living sewage sludge seems to be more dependent on physical activity (such as adsorption and ion exchange) and on chemical interactions (such as chemisorption, complexation, and surface precipitation) between the bacterial surface and the metal ions rather than on metabolic activity, confirming the process to be mainly a passive one (3). Therefore, sewage sludge could be equally applicable as a nonliving system, avoiding many problems (such as the toxic action of metals on living bacteria, the microorganisms feeding, or the negative influence of pH when the pH is outside a certain range, usually 6.5–7.5), associated with a living system. It is worth noting that in spite of the promising results in metals removal, the majority of investigations have examined interactions between living activated sludge and metals. Only limited references exist in which sewage sludge is used as a nonliving, dried, and powdered biosorbent material (10, 11).

Presently, the main methods for sewage sludge disposal include incineration, landspreading, and landfilling; nevertheless, waste microbial biomass may well be an alternative, more economic metal sorbent in comparison with more conventional ones such as activated carbon, alumina, silica gel, etc., but in this case landspreading must be rejected as a final disposal method of metal-laden activated sludge. Landfilling in special landfills and incineration are not influenced by this application.

Preliminary results for the use of nonliving sewage sludge as a biosorbent for metals removal have been recently presented (12). Assuming that the biosorbed amounts of metal ions form a simple surface layer, the Langmuir-type adsorption equation ($1/q_e = 1/X_m + 1/bX_mC$) was used to depict the isotherms and to calculate the constants b and X_m which characterize sorbent–metal interactions. The main parameters investigated included the initial concentration of toxic metals (cadmium, zinc, and nickel, considered as priority pollutants), temperature, influence on biosorption capacity of initial washing (with water) of the activated sludge used, and desorption characteristics. The last two factors, although important for environmental reasons, have usually been overlooked by other researchers. The selective separation of metals from binary and ternary mixtures was also investigated later on, and suggested an innovative recovery route for toxic metals recycling from wastewaters.

EXPERIMENTAL

Nonliving anaerobically digested sewage sludge (denoted hereafter as AS) from the Central Sewage Treatment Plant of Thessaloniki (Greece) was used in bench-scale experiments after sterilization (autoclaving at 393 K for 30 minutes) and drying (under IR lamps). In certain cases AS was initially washed with deionized water to remove the various soluble constituents. AS solubility was found to vary with the pH value of the dispersion, between 14% (at natural pH, around 7) and 40% (at highly acidic pH, around 2.0). The concentrations of the studied toxic metals initially contained in the AS, i.e., before any use, were found (after acid digestion) to be lower than 0.05 mg Cd/g AS (dry weight), lower than 0.05 mg Ni/g AS, and 3.24 mg Zn/g AS.

A hand homogenizer with a 76-μm clearance was used to obtain a dispersion of the biosorbent during batch sorption tests carried out in shaking flasks located in a thermostatic bath. A contact time of 1 hour was applied because it had been found in preliminary experiments to be sufficient for equilibration. The temperature was kept at 293 K unless otherwise stated. The natural pH (7 ± 0.2) of biomass dispersion in deionized water was most often used; the pH was otherwise adjusted to the desired value and controlled throughout the process by the addition of NaOH or HNO_3 solutions. The initial concentration of AS in the suspension was 1 g/L. Most experiments were performed at least twice, and the average values are presented in the figures when they showed a lower than 3% difference.

Adsorption isotherms were obtained for single-component metal solutions using initial metal ion concentrations between 2 and 150 mg/L. The multicomponent metal solutions Zn/Cd, Cd/Ni, Zn/Ni, and Zn/Cd/Ni, were selected for further studies. They contained initial concentrations of 50 mg/L for zinc and 10 mg/L for cadmium and for nickel, which represent the average concentrations of these metals in actual wastewaters. Stock metal solutions were produced from the respective nitrate salts (analytical grade). Following biosorption, centrifugation was used as a convenient solid (biosorbent)/liquid laboratory separation method. Standard flame atomic absorption spectrophotometry procedures (Perkin-Elmer 2380) were used for chemical analysis of the clarified solutions to determine the residual concentrations of metals.

The quantity q_e (amount of metal sorbed per unit mass of AS–taking into consideration the different AS solubilities) was plotted against C (concentration of free soluble metal at equilibrium) and the empirical constants X_m and b, characterizing the AS–metal interactions, were calculated from the plots. In all other cases the results are expressed as percent of removed metal (Re%). To describe the effectiveness of biosorption as a separation process, a selectivity factor (f), similar to the one used in ion exchange or solvent extraction processes, was introduced for certain occasions. It is given by the formula (when considering two metals, A and B)

$$f = \frac{(C_A/C_B)_{\text{biosorbed}}}{(C_A/C_B)_{\text{remaining}}}$$

The influence of pH on metal desorption from loaded biomass was also investigated by applying a contact time of 1 hour under constant agitation for 1 g/L metal-loaded biomass, which was found to contain 33.35 mg Cd/g AS, 22.12 mg Zn/g AS, or 11.97 mg Ni/g AS after preliminary equilibration with the metal solution.

RESULTS AND DISCUSSION

Biosorption of Toxic Metals

The sorption isotherms of the studied metals are presented in Fig. 1. The sorption of metal ions onto AS is considered to be rather complicated, and usually consists of more than one mechanism (adsorption, ion exchange, complexation, surface precipitation, etc.). The role of surface adsorption during removal of toxic metals by living activated sludge, although rather well-established, is not well-defined regarding the specific applied mechanisms (9). Langmuir isotherms of the L-type were found to be the most adaptive to the observed results according to the general classification of adsorption isotherms from aqueous solutions (13). The

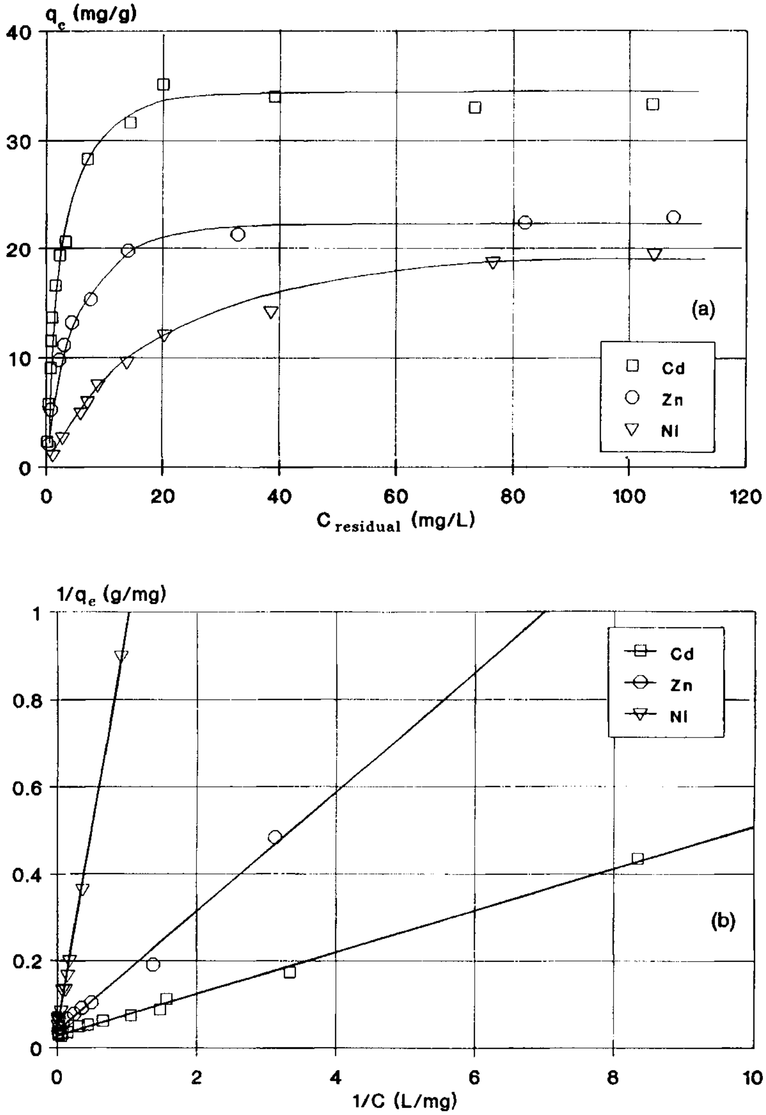

FIG. 1 (a) Sorption of cadmium, zinc, and nickel onto (unwashed) AS; (b) the respective Langmuir isotherms; pH natural (7 ± 0.2), temperature 293 K.

Freundlich equation does not fit as well. This could strengthen the assumption of monolayer adsorption and may give some evidence of the adsorption mechanisms, but it has to be stressed that although a particular isotherm may fit a curve, this is not necessarily of any physical significance (2).

As detailed elsewhere (14), the main assumptions on which Langmuir isotherms are based include: (a) the surface of the sorbent has a certain number of binding sites per surface unit and each one can adsorb only one specific ion, considered to be noninteractive with the other adsorbed ions. When all sites have been occupied, no further adsorption is possible (which means a monolayer is formed). (b) All sites are equivalent, and the energy of adsorption has to be the same, independent of the ions. In a complicated matrix such as AS, it is rather unlikely that all these conditions are met, and therefore the Langmuir isotherms are considered as relatively poor predictors of natural behavior. It was also reported that S-shaped curves, obtained when studying metal-living sludge interactions, indicate a multilayer adsorption at higher metal concentrations (2). These curves were observed when the solute is strongly adsorbed. There is a strong intermolecular attraction within the adsorbed layer, and the adsorbate is monofunctional, i.e., has a single strong point of attachment.

The L-shaped isotherm, which probably occurs in the majority of adsorption cases from dilute solutions, also indicates a reduction of available binding sites as the metal ion concentration increases. Consequently, the proportion of total metal bound decreases at higher concentrations. Nevertheless, on certain occasions (using living bacteria) both Langmuir and Freundlich isotherms were found to fit the biosorption data sufficiently well (15).

The empirical constants of the Langmuir isotherms for the used (unwashed) AS were calculated for single-component metal solutions. The correlation coefficients (r^2) at an order of 0.99 were found for all cases:

Cadmium: $X_m = 36.54$ mg/g, $b = 0.57$ L/mg
Zinc: $X_m = 25.63$ mg/g, $b = 0.28$ L/mg
Nickel: $X_m = 21.66$ mg/g, $b = 0.05$ L/mg

The following order of binding capacity (and also of binding strength) is suggested: Cd > Zn > Ni, while a different sequence (Zn > Cd > Ni) was reported for biosorption experiments when using living activated sludge (1). It is worth noting also that even though the value of X_m measures the binding capacity of the sorbent surface and that of b quantifies the relative strength of the binding interactions, sludges showing high binding strengths for particular metal ions do not necessarily possess increased capacities. The selectivity orders for the removal of metal ions by several sorbents have been explained by suggesting physicochemical factors, such as the electronegativity of the ions, i.e., ions which possess the greatest ionic potential are more strongly attracted to the sorbent, while for ions of equal charge (bivalent in the present case) the molecular size (hydrated radius) may determine the order of preference for adsorption. As these explanations were provided for different experimental conditions, an interpretation of them to explain other metal sorption cases does not seem to be directly feasible.

Moreover, adsorption of metal ions onto a bacterial surface may be conceptually accepted as the formation of a surface–metal complex ($S + M \Leftrightarrow SM$) with a conditional equilibrium constant K_a (L/g) given by the formula $K_a = [SM]/[M]\{S\}$, where M denotes the free metal species, S the unoccupied surface sites, SM the sludge–metal surface complexes, [] the concentration in mol/L, and {} the concentration in g/L. The K_a value provides a measure of the overall affinity, which means a combination of the number of available sites and the binding intensity (incorporation of X_m and b). Values of K_a can be easily calculated when the Langmuir isotherm is derived from adsorption experiments (4). When applied to complexation, the aforementioned assumptions for Langmuir isotherms may be translated as: (a) the formation of complexes with 1 : 1 stoichiometry, and (b) the identical nature of all binding sites.

Conditional stability constants for the AS used had the following values (the same selectivity order was depicted), noting that the lower removal results obtained in the present case for nickel may be promising from the separation point of view:

Cadmium: $K_a = 20.86$ L/g
Zinc: $K_a = 7.29$ L/g
Nickel: $K_a = 1.08$ L/g

It was also suggested that weakly acidic functional groups were mainly involved in metal cations binding to anaerobically digested AS (16); the equilibrium between bound and soluble metal ions is highly affected by the pH value of AS dispersion and the competition existing between hydrogen and metal cations for these binding sites, possibly suggests an ion-exchange mechanism. In fact, zeta-potential measurements of the AS studied, reported as a function of pH, showed negative values, indicating a negatively charged character with a point of zero charge at an acidic pH of 2.3 (12).

The effect of temperature on metals removal by biosorption was also examined, and the results are presented in Fig. 2. These experiments, like

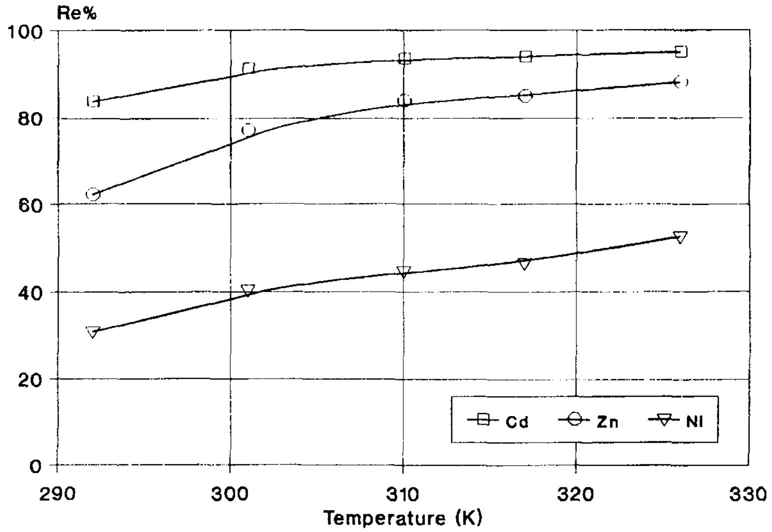

FIG. 2 Effect of temperature on metals removal by biosorption (initial concentrations of each metal 20 mg/L); pH natural (7 ± 0.2).

the previous ones, were carried out independently for each metal. A slight increase in the amounts of biosorbed metals becomes evident by increasing the temperature, mainly in the 293 to 308 K range. This is an indication that the binding mechanism in the cases studied is more likely chemisorption or complexation than simple physical adsorption, or at least there is a complex mechanism dominated by chemisorption. In another case (5), the complexation/removal of the toxic metals Cr, Pb, and Zn, by the use of living AS was found to be independent of the temperature.

Lower removals were observed for nickel than for the other metals, in agreement with other literature findings (5). The strength and nature of binding of toxic metal ions to high molecular weight extracellular biopolymers of AS have been investigated, and nickel was found to be associated predominantly with the soluble fraction of the polymers (17). The stronger affinity of nickel for soluble biopolymers than for biomass could explain the lower binding capacity of AS for nickel, and indicates that these soluble complexes may be unavailable for uptake by the biomass.

In an attempt to improve nickel removal, AS was initially washed with deionized water to remove the soluble constituents before contact of the AS dispersion with nickel solutions. In this case, and in contrast with previous findings, the data were a better fit to the Freundlich model of adsorption ($q_e = KC^{1/n}$, where K and n are empirical constants) rather than to the Langmuir model. Calculation of the empirical constants gave the following values: $K = 4.30$ and $n = 2.76$. Nickel removal was increased up to 75% for natural pH values (7 ± 0.2) and over 95% for pH values above 9, where coprecipitation of nickel hydroxide also occurred (Fig. 3).

With new biosolids disposal regulations in place, the presence of higher concentrations of metals in biosolids could create management problems. Therefore, the examination of toxic metals elution from the AS used is important for a number of reasons, including the stability of the metal-loaded AS and the possibility for either safe disposal in a toxic wastes landfill or for metals recovery and biomass reuse. Toxic metals present or loaded in sewage sludges can be leached out by acid addition and by microbial leaching using iron or sulfur oxidizing bacteria (18).

The desorption results presented in Fig. 4 involve changing the initial pH of the AS dispersion. They show clearly that in an acidic pH range (approximately under pH 5.5), a significant percentage of all the sorbed metals can be released. The behavior of nickel again seems to differ compared with the other metals. It desorbs easier, a fact which may be attributed to weaker interactions with AS and is a further indication of the

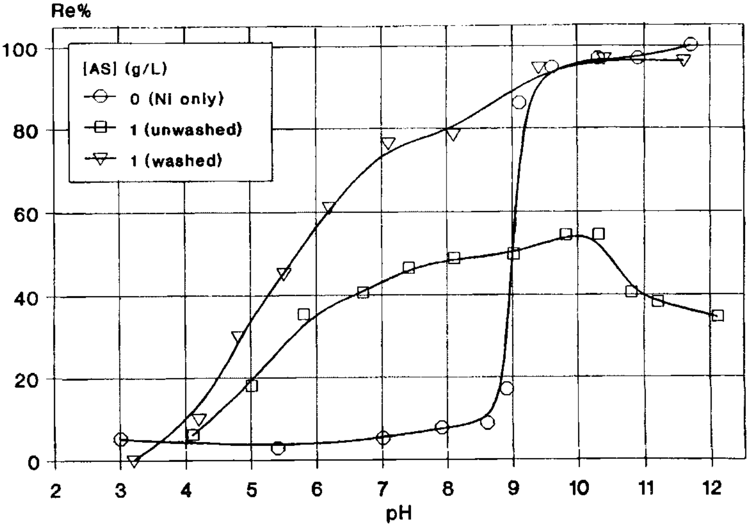

FIG. 3 Effect of dispersion pH on nickel removal (initial concentration 10 mg/L); comparison between washed and unwashed AS.

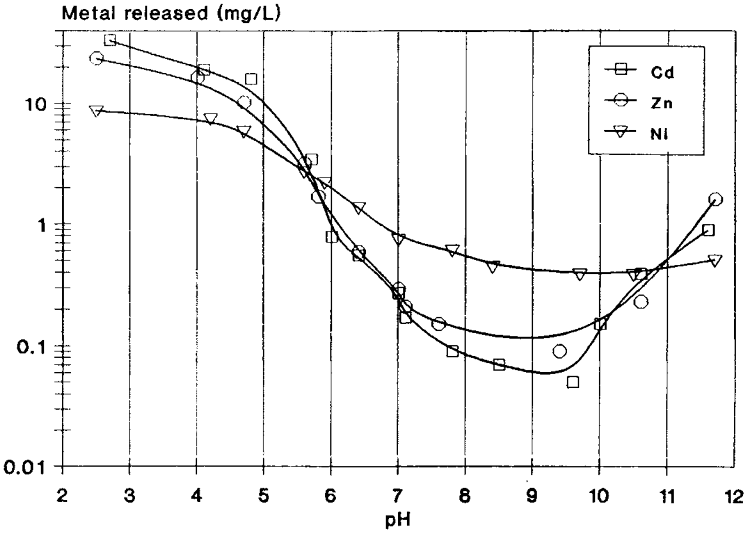

FIG. 4 Desorption study of metals from metal-loaded biomass as a function of dispersion pH.

physical adsorption noticed for this metal. It is worth noting that the relatively easy biomass elution only by pH adjustment could lead to metals recovery in a more concentrated solution, thereby permitting the economical application of other recovery methods, such as electrolysis, as well as the reuse of biosorbent for another cycle of metal removal.

Multicomponent Mixtures

Since AS is a cheap biosorbent material, binary and ternary mixtures of the three studied metals were subsequently examined to check the possible selectivity of the sorbent. The results for the zinc/cadmium binary mixture are shown in Figs. 5 and 6. The different initial metal concentration occurring in most actual cases of effluents is specifically stressed because it directly affects the expression applied for metal removal (Re%). As might be expected, the increase of biosorbent addition in the suspension (up to 3 g/L) produced higher removals for both metals (Fig. 5a), while at 1 g/L AS concentration a decrease in removal for both metals was observed when the initial zinc content was increased (Fig. 5b).

The influence of the dispersion pH for the same Zn/Cd mixture was found to be rather crucial, as shown in Fig. 6. Metal removals by biosorption were observed to increase substantial around a pH value of 5, reached

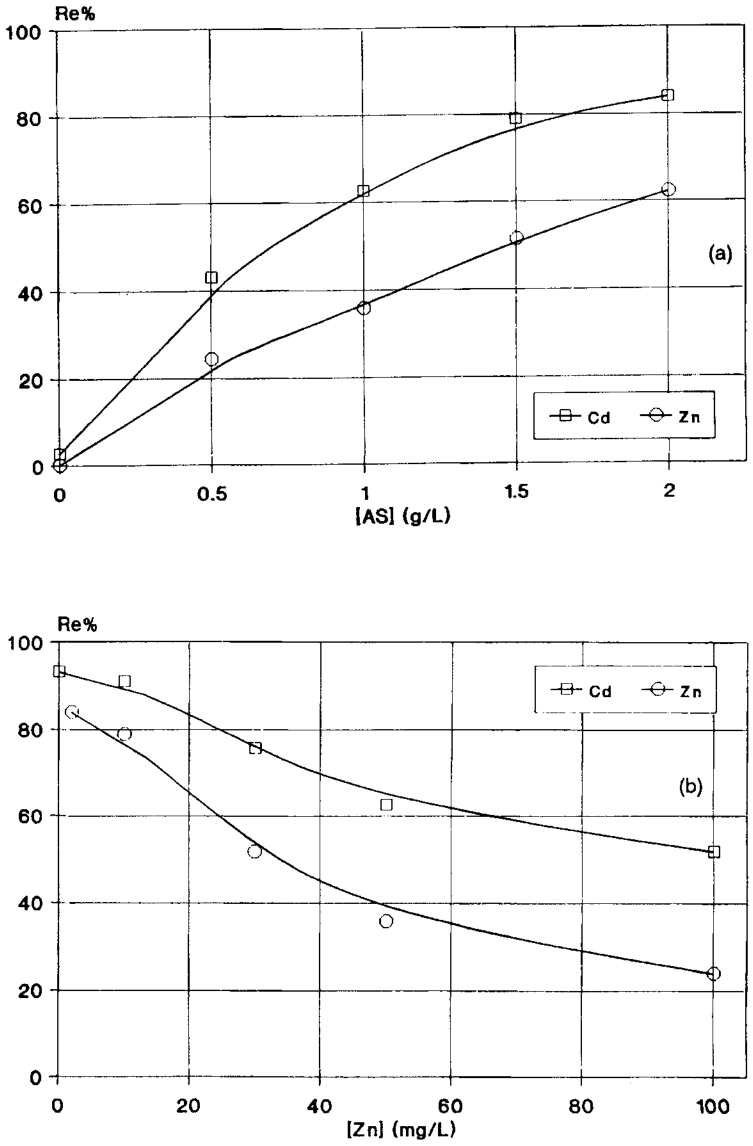

FIG. 5 Zinc/cadmium mixture: (a) influence of AS feed concentration for [Cd] 10 mg/L and [Zn] 50 mg/L; (b) influence of zinc initial concentration for [Cd] 10 mg/L and [AS] 1 g/L; pH natural (7 ± 0.2) and temperature 293 K.

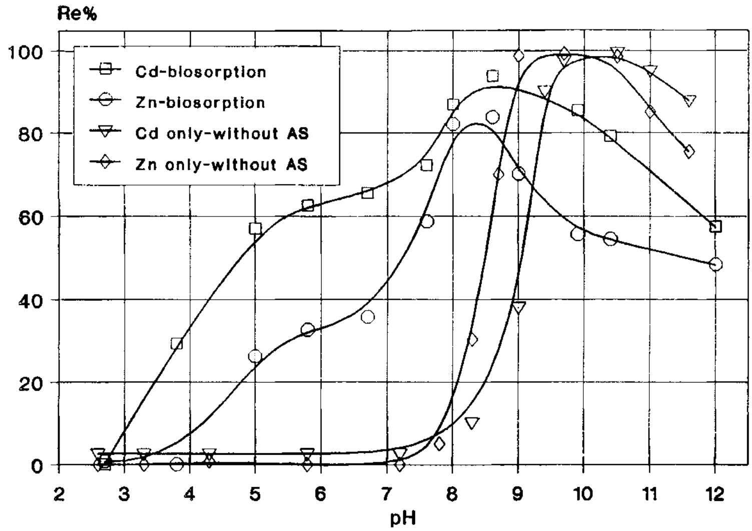

FIG. 6 Zinc/cadmium mixture: effect of dispersion pH on metals removal in the presence and absence of AS.

the highest values (over 80–90%) at pH 8–9, but decreased at highly alkaline conditions (over a pH value of 10) owing to the possible resolubilization of the sorbed metal species. In the same figure the respective experiments for the removal of metals as insoluble hydroxides, i.e., without any biosorbent present, which is the most common process applied for metals removal from wastewaters, are presented for comparison reasons, and they show the highest removals (over 95%) at pH 9.5–10.5 for both metals, where these metals are precipitated. AS particles are capable of binding soluble metal species, as described previously, but they also help to remove insoluble constituents by trapping the formed precipitates, as reported elsewhere (9).

When the zinc/nickel mixture was examined (Fig. 7), the removal of both metals was improved compared with the case where the experiments were performed individually (17), suggesting that nickel adsorbed onto the zinc precipitate and was co-removed. By comparing the biosorption results of Figs. 6 and 7, it can be seen that zinc removals at pH values over 8 and in the presence of cadmium decrease slightly, while in the presence of nickel they increase owing to different interactions between the co-existed toxic metals as well as to different binding affinities.

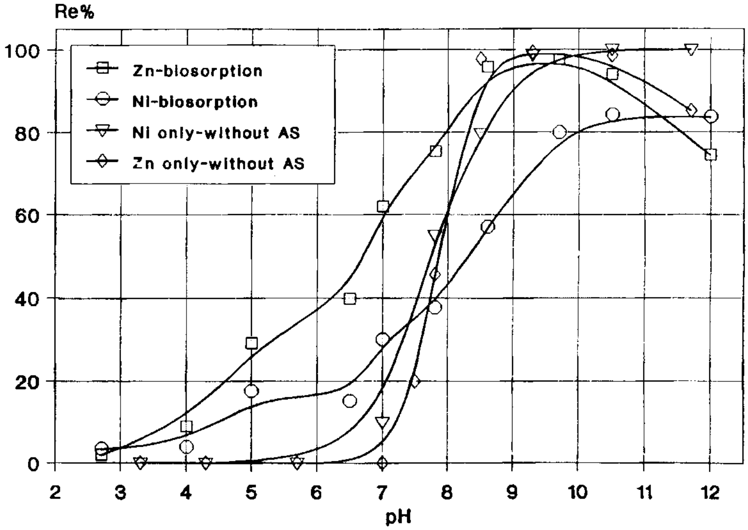

FIG. 7 Zinc/nickel mixture: effect of dispersion pH on metals removal in the presence and absence of AS.

The cadmium/nickel mixture was also examined, and the results of Figs. 8–10 were obtained. Note that equal metal concentrations (10 mg/L of each metal) were used in this case. The influence of AS addition, which varied up to 1.5 g/L (Fig. 8), was found to be the same as in Fig. 5(a), i.e., nickel removal increased with increasing AS content. When the selectivity factor (*f*) was calculated, it was found to reach a value of 20, but it has to be pointed out that selectivity may be highly influenced by the relative concentrations of the various metals present. Additionally, some degree of selectivity might be achieved by microbial strain selection when living biomass is used, as well as in aqueous systems where the metal ions have widely different affinities for the specific biosorbent. Cadmium removals were not influenced by changing the initial nickel concentration between 2 to 20 mg/L, but always remained over 90% (Fig. 9). Similar results were observed for the influence of solution pH (Fig. 10) as in the previously examined mixture of zinc/cadmium (Fig. 6). Moreover, some selective separation was apparent in this case because the selectivity factor *f* reach a value of around 40 in a pH range of 8 to 9.

The ternary mixture (Zn/Cd/Ni) was also examined, and the results are presented in Figs. 11 and 12. It is interesting to note that when the nickel concentration was raised to 30 mg/L, almost no differences in the respec-

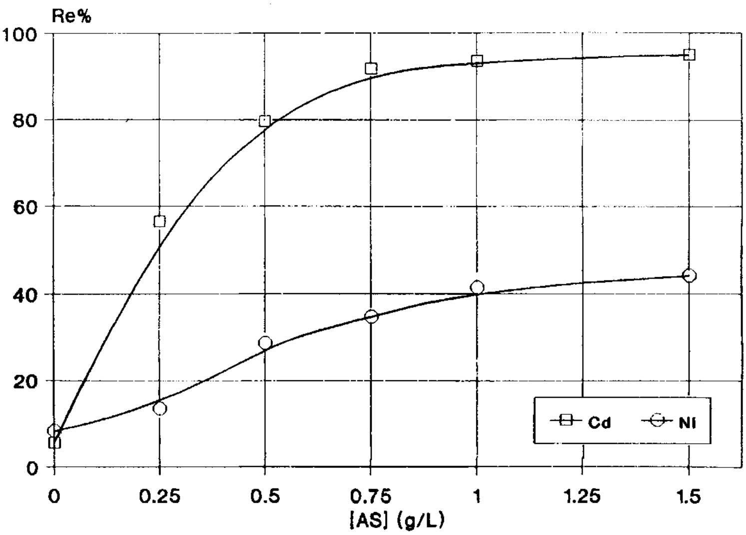

FIG. 8 Cadmium/nickel mixture: influence of AS initial concentration.

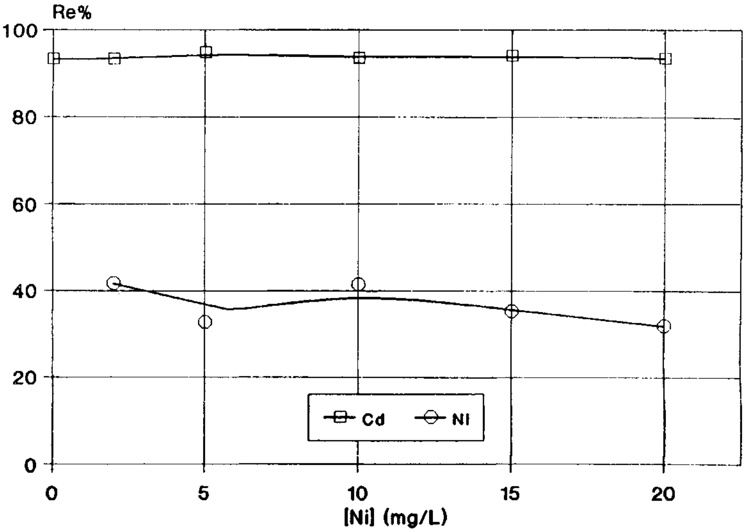

FIG. 9 Cadmium/nickel mixture: effect of nickel initial concentration.

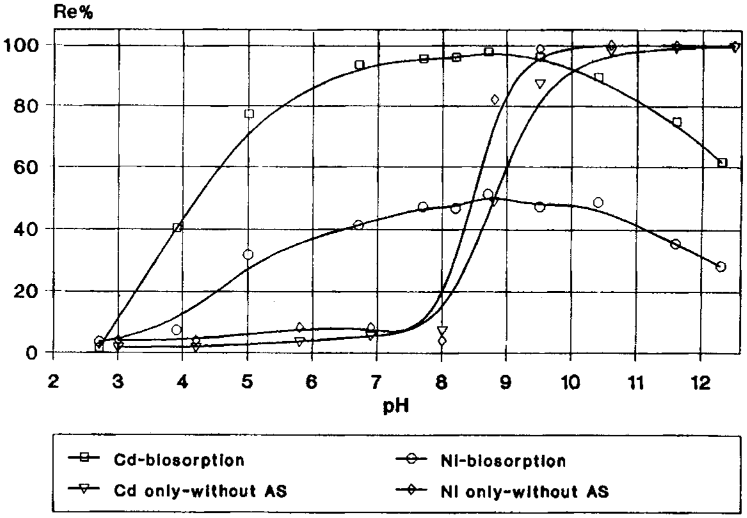

FIG. 10 Cadmium/nickel mixture: effect of dispersion pH on metals removal in the presence and absence of AS.

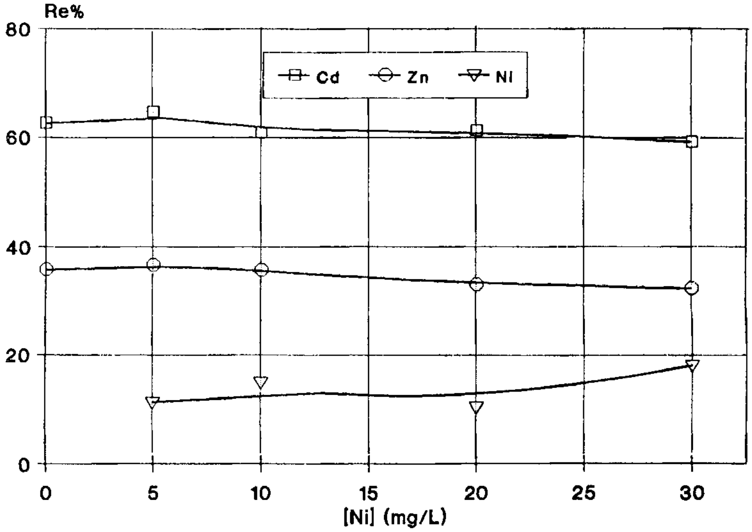

FIG. 11 Zinc/cadmium/nickel mixture: effect of initial nickel concentration.

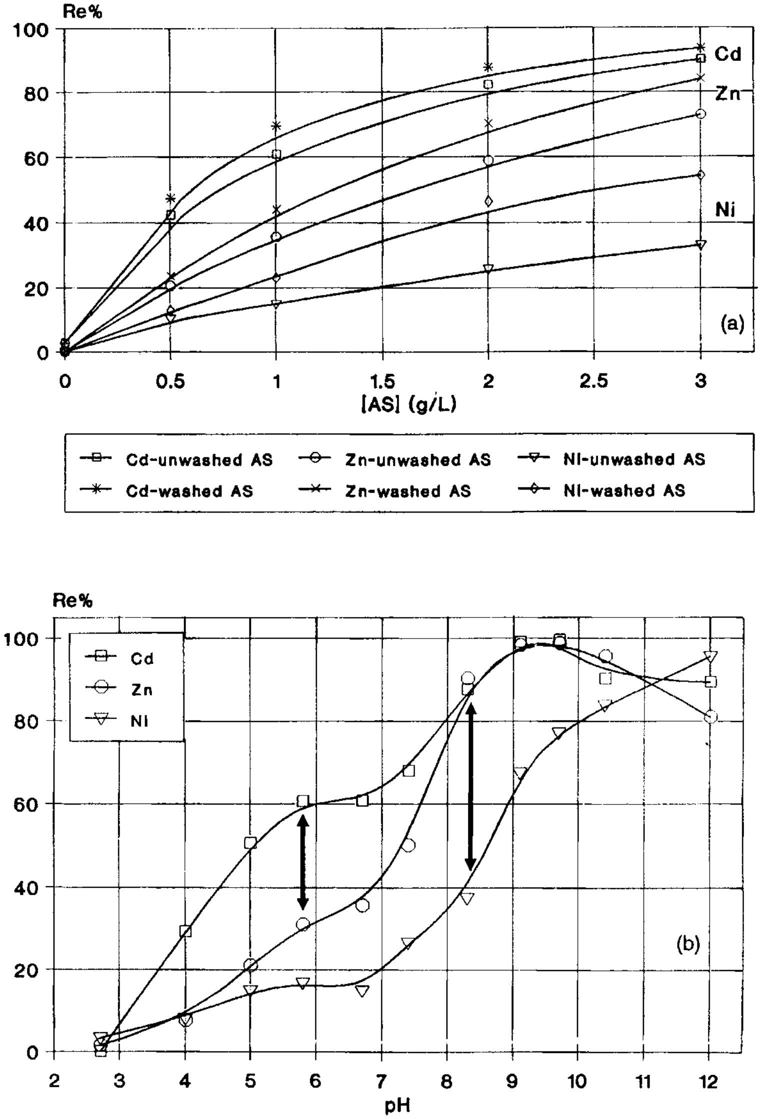

FIG. 12 Zinc/cadmium/nickel mixture: (a) effect of AS initial concentration using washed and unwashed AS at pH 7; (b) influence of dispersion pH on metals removal using 1 g/L AS.

tive cadmium and zinc removals were observed (Fig. 11). Experiments with washed AS (Fig. 12a) showed higher removals for all the toxic metals examined in comparison with unwashed AS. Indications of metal selectivity may be seen in Fig. 12(b) where the influence of pH was investigated. More specifically, in the pH range 5 to 6 between cadmium and zinc and in the pH range 8 to 9 between cadmium/zinc (90% removal) and nickel (around 50% removal). In the case of the ternary mixture, an improvement of zinc and nickel removal was also noted, as previously explained for the results of Fig. 7.

CONCLUSIONS

A number of criteria need to be satisfied if a metal removal/recovery process involving surface binding to microorganisms is to be commercially viable. In evaluating these criteria, it was realized that the examined nonliving (unwashed) anaerobically digested sewage sludge fulfilled most of them.

1. The biosorbent was applied after sterilization and drying but without any other preliminary treatment (activation, etc.).
2. The process seems to be economically feasible because it uses a waste microbial biomass for the treatment of other liquid wastes. A related commercial product has recently appeared (19).
3. High removal efficiencies were obtained for cadmium and zinc (approaching 99% in certain cases), while a preliminary washing step seems to be necessary to remove nickel efficiently. The Langmuir isotherm model was followed by the metal biosorption data.
4. The process was successfully applied over a wide pH range and compared well with the simple precipitation of these metals.
5. The sorbed metals could be rather easily eluted by acidic leaching, resulting in more concentrated metal solutions and making possible the subsequent recovery and reuse of metals as well as of the used biomass. Although this has been not tested for the specific biosorbent, it has been examined for related cases (8).
6. The presence of metal mixtures creates competition for available adsorption sites on the adsorbent surface. Despite the competition, the total adsorption capacity was found to increase, even though the adsorption of a single ion (nickel) was lower.
7. The order of metal capacity (and affinity) for nonliving and unwashed AS was Cd > Zn > Ni.
8. On certain occasions a promising selectivity was noticed among the metals examined when they co-existed in binary or ternary mixtures.

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