

BIODEGRADABLE CHELANTS FOR HEAVY METAL REMOVAL FROM SLUDGE AND SOIL-SLUDGE MIXTURES

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Heavy metal extraction study was performed using five chelating agents with different biodegradability: EDTA, EDDS, MGDA, SCLC and citric acid. Aminopolycarboxylic acids EDDS and MGDA showed high heavy metal extraction capability from sewage sludge and sludge-soil mixtures. According to heavy metal removal efficiency investigated chelants can be ranked in the following order: EDTA≈EDDS≈MGDA>CA>SCLC. Extraction efficiency from sludge mixtures with clay soil was markedly lower than from the mixtures with sandy soil. Biodegradability of EDTA, MGDA, EDDS and CA was evaluated measuring BOD in water environment over 28 day period. According to the results of biodegradability test after 28 days the ranking order of the chelants was following: EDDS (99%)>CA (32.4%)>MGDA (29.2%)>EDTA (14.9%). Results showed that significantly easier than EDTA biodegradable chelating agents, such as EDDS and MGDA, can be successfully used for heavy metal removal from sewage sludge as well as metal-contaminated soil.

Keywords: heavy metals, removal, sewage sludge, chelants, biodegradability

INTRODUCTION

Treatment and disposal of sewage sludge (SS) originated at waste water treatment plants (WWTP) of big cities and industrial areas are important problems world-wide. In Lithuania thousands of tons of SS, which accumulated over the years, are stored in special sludge storage sites and, if not handled properly, can pose threat to surface water, groundwater, soil, plants and animals as well as human beings (Praspaliauskas and Pedišius, 2017). Recycling of the sludge is a preferred solution for sustainable development, rather than the incineration or disposal. National Waste Management Plan of the Republic of Lithuania for the period of 2014-2020 (LR valstybinis, 2014) specifies that the disposal of SS in municipal landfills, sludge storage sites or other long-term storage facilities shall be terminated by the establishment of appropriate regional sludge management capacities. Unfortunately, in the case of smaller WWTP, sludge is still disposed at storage sites, sometimes even without dewatering, or site itself not properly constructed (Feasibility study, 2018).

There is no common practice in the world for SS management – sludge can be incinerated with or without energy recovery, dumped in the landfills, disposed at seas or stored in special storage facilities (Dargenis et al., 2008). Another important alternative is SS recycling using the valuable properties of the sludge. Sludge on-land application can effectively improve soil properties such as pH, organic matter and plant nutrient content, cation exchange capacity, bulk density, water retention, porosity and aggregate stability, and finally enhance plant growth and increase the crop yields (Wei and Liu, 2005; Epstein, 2003; Korentajer, 1991). High amount of organic matter in SS also have a positive effect on soil physical properties, improving soil structure, increasing soil aeration, and maintaining optimal soil moisture (McGrath et al., 2000; Shchanchar et al., 2000). However on-land sludge application is often limited by elevated concentrations of heavy metals (HM), which can have negative impact on the environment and human health (Alloway, 2013; Kabata-Pendias, 2011; Ministry of, 2001). This impact can be effectively reduced by removing HM from the sludge, when different chemicals are used for metal extraction from the solid substrate: as e.g., aqueous solutions of mineral acids and their salts, organic acids, oxidizing/reducing agents as well as organic compounds creating organic complexes with HM (Bolan et al., 2014; Komarek et al., 2007; Poletini et al., 2007; Zaleckas et al., 2007; Yuan et al., 2007; Zhang et al., 2006; Moutsatsou et al., 2006; Khodadoust et al., 2005; Kos et al., 2004; Lee et al., 2004; Marchioretto et al., 2002; Sun et al., 2001). Unfortunately, despite of high efficiency, but due to their poor biodegradability stable organic compounds used for extraction can persist in the environment for many years, causing serious environmental problems (Renella et al., 2004; Kos and Lestan, 2003; Grčman et al., 2003; Bucheli and Egli, 2001; Takahashi et al., 1997). Primary biodegradation – is the alteration in the chemical structure of a substance, brought about by biological action, resulting in the loss of a specific property of that substance. While ultimate biodegradation (aerobic) – is the level of degradation achieved when the test compound is totally utilised by micro-organisms resulting in the production of carbon dioxide, water, mineral salts and new microbial cellular constituents (biomass). Chemical compound is defined as readily biodegradable after passing certain

specified screening tests for ultimate biodegradability: it is assumed that such compound will rapidly and completely biodegrade in aquatic environments under aerobic conditions (OECD guidelines, 1992).

The aim of this research was to carry out a comparative study of biological degradation of the most effective chelating agents that can be used for heavy metal removal from the solids.

RESEARCH METHODS

Heavy metal removal. Chelant induced heavy metal extraction has been performed with strong chelating agents – 5 different aminopolycarboxylic acids. Chemical name, abbreviation and structural formula of the selected chelants, indicating the number of carboxyl groups, are given in Table 1. Removal tests were carried out with SS, originating from Kaunas WWTP, and two different sludge-soil mixtures: SS with clay soil and SS with sandy soil. Extractions were performed in a shaker with 0.1M aqueous solutions of chelating agents (pH 6), at a solid/liquid ratio 1:10, and room temperature (20°C). All extractants used were of analytical reagent grade.

Table 1. Chelating agents used for heavy metal extraction

Extracting agent (Abbreviation)	Number of carboxyl groups	Chemical structure
Citric acid (CA)	Tricarboxylic acid	
Methyleneglycinediacetic acid (MGDA)	Tricarboxylic acid	
Ethylenediaminetetraacetic acid (EDTA)	Tetracarboxylic acid	
S, S'-ethylenediaminedisuccinic acid (EDDS)	Tetracarboxylic acid	
S-carboxyl-L-cysteine (SCLC)	Dicarboxylic acid	

EDTA was used as trisodium salt Na₃EDTA. EDDS was chosen as the S,S-isomer of EDDS, because the R,R- and S,R-isomers are not so easy biodegradable (Schowanek et al., 1997; Hauser et al., 2005).

Chelant biodegradability. Biological degradation of chelating agent in an aqueous media was determined by measuring oxygen consumption in a closed respirometer according OECD (Organisation for Economic Co-operation and Development) guidelines for testing of chemicals using 301 D Closed Bottle method (OECD guidelines, 1992). For this method, the test substance should be non-volatile and have solubility in water of at least 100 mg/l. Moreover, at least empirical formula of the substance and its purity, or relative proportions of major components, should be known. Elemental composition is obligatory so that theoretical oxygen demand (ThOD) may be calculated. In case the ThOD cannot be calculated, chemical oxygen demand (COD) should be determined, but falsely high values of percentage biodegradation may be obtained if the test substance is incompletely oxidised in the COD test. Insoluble and volatile substances may be assessed provided that precautions are taken. Degradation values for insoluble substances may be falsely low unless the bottles are agitated periodically during the incubation. This method allows a maximum of 6 month period for the determination of biodegradability. The solution of the test substance in mineral medium is inoculated with a relatively small number of micro-organisms from a mixed population and kept in completely full, closed bottles in the dark place at constant temperature. Light is excluded seeking to avoid growth of algae in the bottles during incubation. Degradation is followed by analysis of dissolved oxygen over a 28 days period. The amount of oxygen taken up by the microbial population during biodegradation of the test substance, corrected for uptake by the blank inoculum run in parallel, is expressed as a percentage of ThOD or, less satisfactorily COD. Modifications according to the methodology described in a normative document LAND 47-1 (2002) were also made for the determination of biochemical oxygen demand (BOD) in water samples. The composition of the aqueous medium was made following the steps described in the methodology OECD, 301 D Closed Bottle. Solutions for mineral medium were prepared using analytical grade reagents: solution (1) – 8.50 g of potassium dihydrogen phosphate (KH₂PO₄), 21.75 g of dipotassium hydrogen phosphate (K₂HPO₄), 33.40 g of disodium hydrogen phosphate dihydrate (Na₂HPO₄·2H₂O) and 0.50 g of ammonium chloride (NH₄Cl) was dissolved in deionised water and made up to 1 L and pH 7.4; solution (2) – 36.40 g of calcium chloride dihydrate (CaCl₂·2H₂O) was dissolved in water and made up to 1 L; solution (3) – 22.50 g of magnesium sulphate heptahydrate (MgSO₄·7H₂O) was dissolved in water and made up to 1 L; solution (4) – 0.25 g of iron (III) chloride hexahydrate (FeCl₃·6H₂O) and 0.4 g of disodium salt of ethylene-diamine-tetraacetic acid (Na₂EDTA) was dissolved in water and made up to 1 L. Preparation of mineral medium was made by mixing 1 mL of each solution (1), (2), (3), (4)

with 800 mL of deionised water and then made up to 1 litre. Solutions containing test substances were prepared by adding the chemical (test or reference substance) directly to the mineral medium, as all tested chelants were characterised by good solubility in aqueous neutral solutions. The inoculum was prepared using commercially available inoculants – biological tablets containing microorganisms that break down fat, paper and other organic materials. Chelant concentration in incubation vessel was 100 mg/L. Oxygen concentration in incubation vessels was measured over a period of 28 days. Dissolved oxygen concentration was determined electrochemically using a calibrated OXI 597 oxygen meter. During the preparation of test bottles mineral medium was strongly aerated for at least 20 minutes and allowed to stand still. After standing for 20 h the medium was ready for use at the test temperature. Each test series was carried out with mineral medium derived from the same batch. A sufficient number of BOD bottles, including inoculum blanks, was used to allow triplicate measurements of oxygen consumption to be made at the desired test intervals. In order to ensure that the inoculum activity is not limited, the concentration of dissolved oxygen in the BOD bottles must not fall below 0.5 mg/L. Replicate bottles were completely filled with no air spaces, then stoppered ensuring that no air bubbles are enclosed, and incubated at 20°C in the dark. Each series must be accompanied by a complete parallel series for the determination of the inoculated blank medium.

RESEARCH RESULTS

EDTA is a widely tested and strong chelating agent, and there is no doubt about its effectiveness. It is known that EDTA shows excellent results in removal metals from soils and sediments, but unfortunately, due to its poor biodegradability, it is also very persistent in the environment. This can cause possible long-term effects on speciation and bioavailability of toxic as well as essential trace elements (Labanowski et al., 2007; Cao et al., 2008). Moreover, surface water and ground water pollution can be activated by leaching of persistent chelants and their metal compounds (Takahashi et al., 1997; Schowanek et al., 1997). These are the main reasons impeding the application of EDTA as well as other persistent chelating agents for heavy metal removal from the solids. Therefore, presuming that EDTA is not the best choice for soil washing, comparable tests were carried out with other aminopolycarboxylic acids. HM extraction experiment was performed using five different chelating agents: EDTA, EDDS, MGDA, CA and SCLC. Results showed that complexation efficiency was strongly dependent upon the investigated metals and chelants applied, but according to their best performance, the investigated chelants can be ranked in the following order: EDTA ≈ EDDS ≈ MGDA > CA > SCLC. Meanwhile, the ranking order of the heavy metals according to the generalised extraction efficiency was following: Zn > Cu > Ni ≥ Pb > Cd >> Cr. Overall HM removal capacity of EDDS was better than that of MGDA in all cases, except for Cr. Extraction efficiency of all HM from SS mixture with clay soil was markedly lower than that from SS mixture with sandy soil. These results confirm lower HM mobility and availability in clay soil than in sandy soil due to specific immobilising effect of clay minerals. Extraction efficiency of SCLC was the lowest from the tested chelants.

Rapid biodegradation could be a priority for a chelating agent to be used in environmental technology. Despite of its high extraction efficiency, EDTA is not a desired chemical when released into the environment. High chemical stability of this synthetic chelant is then considered as disadvantage. EDTA has relatively low biodegradation rate both in soil and water environment (Bucheli-Witschel and Egli, 2001). This can lead to HM leaching and contamination of ground water and surface water (Nowack, 2002). According to OECD guidelines (1992) theoretical oxygen demand is the calculated oxygen amount required to oxidize a test organic chemical to final oxidation products. Actual oxygen uptake during biodegradation of different organic chemicals is called the biochemical oxygen demand, and it depends on the biodegradability of the chemical compound and the specific microorganism degrading the compound. BOD is measured experimentally, and is describing the amount of biochemically oxidizable material present in the test solution expressed in terms of the oxygen required to break it down. Most often, BOD is expressed in the number of milligrams of oxygen consumed by microorganisms when metabolising 1 mg of test compound during incubation in the dark at certain temperature over a specific time period (OECD guidelines, 1992). During this experiment the biochemical oxygen demand after each time period was calculated by subtracting (1) the oxygen depletion (mg O₂/L) of the inoculum blank from that exhibited by the test substance during chelant biodegradation:

$$BOD = \frac{BOD_x - BOD_0}{C_x} \quad (\text{mg oxygen / mg test substance}) \quad (1)$$

where:

BOD_x – oxygen consumption in mg O₂/L per incubator with chelating agent (uptake by test substance);
 BOD_0 – oxygen consumption in mg O₂/L per incubator without chelating agent (uptake by blank);
 C_x – concentration of chelating agent mg/L in a test vessel.

The chelant biodegradability (BD) in % was calculated using the formula (2):

$$BD = \frac{BOD}{ThOD} \times 100 (\%) \quad (2)$$

where:

BOD – biochemical oxygen demand in mg O₂/mg of a chelating agent (test substance);
 $ThOD$ – theoretical oxygen demand in mg O₂/mg of a chelating agent (test substance).

Theoretical oxygen demand (ThOD) was calculated using test substance elemental composition. For the compound $C_cH_hCl_{cl}N_nNa_{na}O_oP_pS_s$ the ThOD was calculated using the formula (3):

$$ThOD = \frac{16[2c + 0.5(h - cl - 3n) + 3s + 2.5p + 0.5na - o]}{MW} \quad (3)$$

where:

MW – molar weight of chelating agent (g/mol);

c, h, cl, n, s, p, na, o – the number of atoms of a corresponding element in the empiric formula of chelating agent.

Seeking to assess chelant persistency in the environment comparative study of their biodegradability in aqueous media was carried out. Figure 1 presents the dynamics of biological decomposition of the selected chelating agents. Data showed that within 28 days 99% of EDDS was degraded. Meanwhile, EDTA showed the lowest biodegradation rate in aqueous media comparing with the other investigated chelants – EDDS, MGDA and CA.

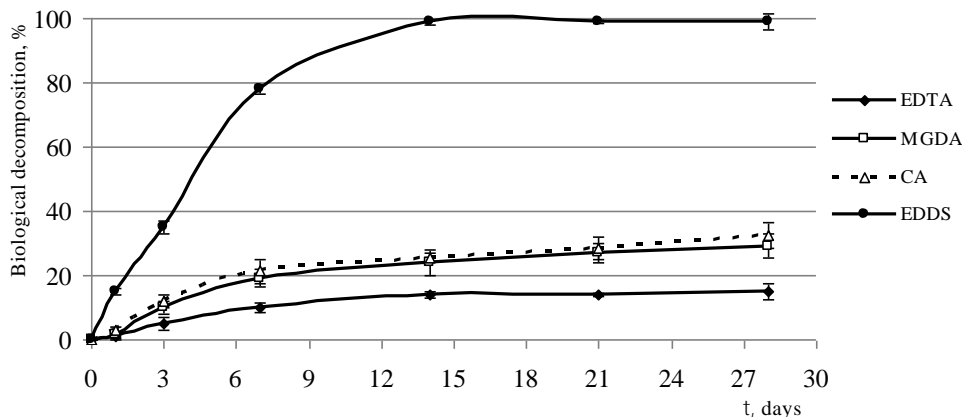


Figure 1. Biological degradation of chelating agents: EDTA - ethylenediaminetetraacetic acid; CA - citric acid; MGDA - methyleneglycinediacetic acid; EDDS - S, S'-ethylenediaminedisuccinic acid

Biological degradation of EDDS reached 98% already after 14 days. Tandy et al. (2006) found that the biodegradation half-life of EDDS in soil is 3.8-7.5 days, depending on the initial chelant concentration. Results obtained in our study indicate that EDDS half-life in water is equal to 4.2 days. A series of other biodegradation studies have been conducted using conventional methods with sewage sludge as inoculant, and complete degradation of this chelant was recorded in a period from 5 to 16 days (Jaworska et al., 1999; Schowanek et al., 1997; Takahashi et al., 1997). According to the biodegradability results obtained during 28-day test of this study, chelant ranking order was following: EDDS (99%) > CA (32.4%) > MGDA (29.2%) > EDTA (14.9%). OECD 301 D standard states that a substance is considered to be readily biodegradable if more than 60% degradation is observed over a 28-day period. Thus EDDS can be defined as easy biodegradable in the environment and can be successfully applied for soil/sludge washing instead of EDTA. EDTA was found to have the lowest biodegradability in aqueous media compared to other chelants used in the study. CA and MGDA do not exhibit high biodegradation rate, but both chelants were significantly easier biodegradable if compared with EDTA.

CONCLUSIONS

- Aminopolycarboxylic acids EDDS and MGDA showed high heavy metal removal capability from sewage sludge and sludge-soil mixtures. According to heavy metal removal efficiencies investigated chelants can be ranked in the following order: EDTA ≈ EDDS ≈ MGDA > CA > SCLC. Extraction efficiency of metal removal from sludge mixture with clay soil was markedly lower than from the mixture with sandy soil.
- According to the biodegradability results after 28 days chelant ranking order was following: EDDS (99%) > CA (32.4%) > MGDA (29.2%) > EDTA (14.9%). Results showed that easier than EDTA biodegradable chelating agents, such as EDDS and MGDA, can be successfully used for heavy metal removal from sewage sludge as well as metal-contaminated soil.

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