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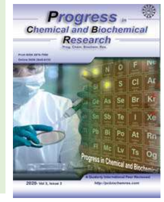

Original Research Article

Investigation of the Process of using Sludge Contaminated in the Effluent of Chemical Industries

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A R T I C L E I N F O

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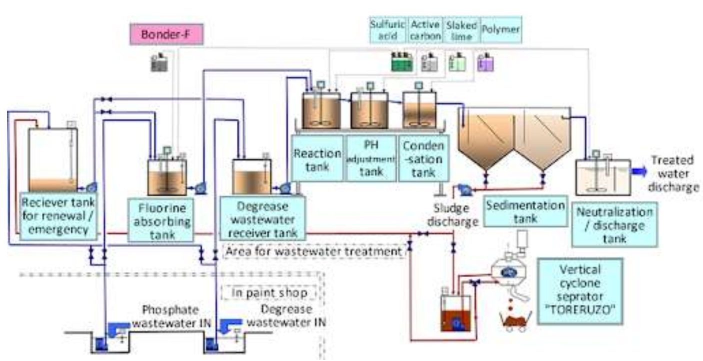
A B S T R A C T

The purpose of this study is to evaluate the effects of increasing sewage sludge as a fertilizer on the stimulation of biological processes and the rate of bioremediation of fatty substances in soils contaminated with this substance in the effluent of chemical companies. Industrial wastewater, especially the chemical industry, is one of the largest volumes of industrial wastewater producers. The chemical industry in general around the world produces more than 1.4 million barrels of contaminated sludge per day, which is a major environmental hazard. If the environment is not considered in the design and operation of industrials, in addition to environmental problems related to air and soil pollution, it will cause surface and groundwater pollution, the scope of which can affect the next few generations. Therefore, several solutions have been proposed for the treatment of contaminated wastes, which are mentioned in this study. Due to limited soil and groundwater resources, soil pollution is one of the most important environmental problems in the world. For this purpose, methods such as direct engineering methods or natural cleaning, i.e. without human intervention in modifying this control, reduce or eliminate pollution from the environment by increasing biological activities of the environment. Also, by using methods that accelerate the decomposition of pollutants such as usage of nitrogen and phosphorus organic matter and plowing for better soil aeration increasing microbial activity is accelerated.

G R A P H I C A L A B S T R A C T



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Introduction

Awareness of the effects of the spread of chemical products in various industries is one of the most important pieces of information that should be available to researchers in environmental studies to combat contamination of aquatic and terrestrial environments. The widespread use of heavy metals in the industry has led to an increase in the production of wastes and effluents in which these metals are found. In many cases, these considering issues cause environmental problems by their excess concentration of pollutants, and as result pollution in various environments, including water, sewage, air, and soil emerged. There are several methods for disposing of sewage sludge, among which land use, incineration, and sanitary burial are the most ones, but if there is heavy metal in the sludge, in none of these cases the sludge is treated. There are several methods for cleaning sludge and even contaminated soils as well as recycling these metals from the environment. These methods include stabilization and solidification methods, washing, thermal and adsorption, flushing, refining, and electrokinetic plants [1]. It should be mentioned that three-strand organisms that cause the foam to form in activated sludge include Nocardia, microtrix paroxysmal, and actinomycetes. This type of foam consists of active sludge solids (flocs) that contain a large number of filamentous bacteria that have grown on the surface of the flocs and form a stable, brown layer in the surface of the final fan and settling pond. Also, Nocardia and microtrix occur with increasing sludge age [2]. As the amount of grease and fat compounds in the sewage increases, so does the amount of effluent pollution produced by Industrials depending on the type of activity and by what devices they are produced, so the wastewater treatment systems and processes that can be used in this industry are very complex and different. Refinery effluent sources include cooling towers, steam generators, and process units, whose effluents are highly hazardous due to their hydrocarbon and aromatic compounds and are considered serious environmental damage. Due to the large and important nature of the industry and the high volume of its effluent, several mechanisms have been developed for the treatment of effluents, which we will briefly explain in the rest of this article [3].

Industrial Wastewater Treatment Processes

Wastewater treatment in various industries, including textile, iron and steel, mining, chemical, and other various chemicals industries are done in four stages: primary, physicochemical (secondary), biological and final processes. In the first step, which is a prelude to facilitating the next steps, the suspended particles, emulsions, and free hydrocarbons are mechanically removed from the effluent. In this stage, large solid or floating materials such as fat compounds, grease, or other compounds are removed from the effluent. In the second stage, which is physicochemical treatment, the first chemicals that have clotting properties are added to the effluent and then large particles become coagulated and settle, the clots and coarse particles are separated from the effluent. In the third stage, which is called bioremediation, by adding oxygenated, aromatic, sulfur compounds; biological compounds are removed from the effluent. In the last step, which aims to meet environmental standards for the effluent to enter nature, more phosphorus compounds are removed [4].

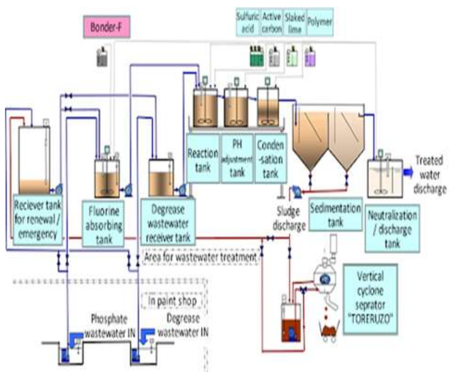

Fig 1. Waste Water Treatment Process

Wastewater Treatment Methods in the chemical Industry

There are several methods for wastewater treatment in the chemical industry, which can be mentioned as followed (Table 1). Also, it is provided a list of the most important technologies used in wastewater treatment of chemical industries (Table 2). On the whole considerable researches carried out in this field of study as followed.

Table 1. wastewater treatment method

wastewater treatment method	Description
API separator	The first and most important treatment step that collects suspended solids in the effluent
DAF process	Used to purify the oil-water emulsion
Electrical coagulation	Use electrical currents to stabilize suspended particles and clot them
Chemical coagulation	Help settle sediments by adding chemicals called coagulants to effluents and reducing the electrical charge of particles
Biological methods	Use microbial metabolisms, aerobic and anaerobic microorganisms to settle and separate solid particles and activated sludge
Membrane separation	The membrane acts as a filter between two phases, water passes through the membrane while suspended solids and other materials are trapped on the other side of the membrane
Nanotechnology	Nanomaterials are easily bonded to contaminants and recycled by magnetic pumps. Due to the very small size of nanoparticles and their high proximity to pollutants, almost 100% of pollutants will be removed.

In 2000, Namkung et al. found in bioremediation of diesel-contaminated soils that mixing contaminated soil with sewage sludge at a ratio of 0.1: 5 showed the highest contaminant degradation [5]. In 2003, Gogi et al. in the bioremediation of Soils contaminated with fatty and hydrocarbon compounds found that aeration, application of nitrogen and phosphorus organic matter, and microbial inoculation degraded 75% of the contaminants.

Table 2. Methods and processes

No.	Process	Wastewater type	Removed contaminants	Removal efficiency
1	Physic-chemical	Wastewater/effluent	AllIndustrial products Aromatic hydrocarbonproducts	16 24
2	Membrane processes	Effluent of refinery	Fatcontent	69
3	MBR	Chemical industryeffluent	Heavy metals Fe	70 75
4	MWCNT with polyvinylidene fluoride pvdf	Effluent of refinery	fat	
5	CF-MBR	Chemical industryeffluent	COD	93
6	HF-MBR	Effluent of refinery	COD BOD TSS VSS Turbidity	82 89 98 99 98
7	Batch membrane reactors	Combined fatwastewater	Hydrocarbon pollutants	97
8	UF	Chemical industryeffluent	COD	44
9	Coagulation separation by Al2Cl3 & Fe2Cl3	Chemical industryeffluent	COD	58
10	Coagulation separation and freezing by PZSS & A-PAM	Heavy fatwastewater	fat	99

Prerequisites for the treatment laboratory

To determine water quality, chemical and microbial water tests are performed, and what is important before testing is accurate sampling in terms of number and repeat sampling. Samples should be tailored to the goals of providing safe drinking water [6].

Required Tests

The number and type of tests required in a water treatment plant depend on the quality of raw water, the treatment process, and a number of treatment units. The type of tests required, along with the reasons for the test, to control the operation of a treatment plant and control the quality of treated water are described below. On the other hand usage of a continuous system is also recommended for measuring many parameters [7].

Physical-Chemical Experiments

Temperature: In general, cold water is better for drinking than hot water. High water temperatures accelerate the growth of microorganisms and are effective in creating flavor, aroma, color ,and corrosion. The standards do not specify any guidelines for water temperature, but the temperature should be acceptable to the consumer. The temperature of the water plays an important role in the coagulation process [8].

Color: Watercolor is usually due to the presence of pigmented organic compounds (mainly humic and fulvic acids), plant decomposition, dissolution of soil in water, as well as minerals such as colloidal iron, manganese ,and other metals, which are formed as natural impurities.

Smell: Excessive activity of aquatic and biological organisms is the main cause of odor in surface water resources. Odor is also mentioned as one of the by-products of disinfection (chlorination). The Smell may be a sign of contamination or defects in water treatment or distribution operations. If the number of odors increases too much, the cause should be investigated immediately. The simplest way to measure odor is to use the TON number or odor threshold number, which is one of the sensory methods [9].

In this method, a 200 ml sample of water is diluted with distilled water at a constant temperature of 40 ˚C so that it no longer smells, then the TON number is obtained from the following equation (Eq. 1).

$$TON = ((\text{sample initial volume} + \text{DM consumed volume})) / \textit{water initial volume} \qquad \text{(eq. 1)}$$

The TON number in the Iranian national standard is 2 units at 12 ˚C and 3 units at 25 ˚C.

Turbidity: Water turbidity is due to the presence of colloidal particles, which are often caused by the increase of suspended solids in the water supply source. This increase is due to atmospheric factors such as rainfall and floods. High levels of turbidity can reduce the effect of disinfectant on microorganisms and increase the growth of bacteria in the distribution network.

Total Suspended Solids (TSS): All suspended solids are in the form of suspended solids in water and generally in the form of glues. Comparison of concentration of these solids obtained from inlet water and outlet water from filters shows the efficiency of units in removing and reducing suspended solids. Most solid suspended particles can be separated from water by filtration; therefore, suspended solids in a water sample can be estimated by filtering water, drying the residual mass, and filtering to a constant weight at temperature, and determining residual mass on the filter. The results of this test show the suspended solids in terms of dry mass in volume (mg/l)[10]. The amount of dissolved solids passing through the filter is also expressed in milligrams per liter; this value is equal to the difference between total solids in the water and suspended solids in a water sample.

pH: Although pH within the standard range does not normally have a health effect on consumers, from an exploitative point of view, it is one of the important parameters of water quality. To achieve optimal treatment and disinfection, careful attention to pH in all stages of water treatment is necessary for effective disinfection with chlorine, it is necessary to adjust the pH range <8. Adjust the pH of water in the saturated pH range in the distribution network for minimum corrosion or fouling is required in the system. High pH values can result from the discharge of industrial effluents into water sources.

*Measuring Oxygen Solution (DO):*The solubility of oxygen in water is a function of the temperature and partial pressure of this gas, which is measured once a day using an oxygen meter from the inlet and outlet of a treatment plant.

Electrical Conductivity (EC): Electrical conductivity is one of the quality indicators of drinking water that is simple and quickly measurable, and expresses the ionic power of water to transmit electricity. Electrical conductivity is measured in micromoles per centimeter and varies with temperature, concentration, and type of water ions.

Total Soluble Solids (TDS): All soluble solids include water-soluble mineral salts in the form of cations (calcium, magnesium, potassium, sodium, iron, manganese, etc.) and anions (bicarbonates, hydroxides, chlorides, sulfates, nitrates, phosphates, etc.). The amount of TDS in water varies greatly due to differences in mineral solubility in different geographical areas. The amount of TDS in water has a significant effect on the taste of drinking water. Water with a TDS of less than 600 mg/L is normally considered good for drinking. The maximum TDS in the World Health Organization standard is 1000 mg/l.

Total Alkalinity (TA): The total alkalinity of water is the number of ions in the water that are neutralized by the reaction with hydrogen ion. Thus, alkalinity is the degree to which water can neutralize acids. These compounds are obtained from the dissolution of soil and atmospheric minerals. Typically, bicarbonate ions, carbonate, and hydroxide form water alkalis. Excessive alkalinity causes the bitter taste of water [11].

*Total hardness (TH):*In general, the concentrations of dissolved calcium and magnesium salts make up the total hardness of water, expressed in terms of calcium carbonate. Depending on pH and alkalinity, hardness of more than 200 mg/l due to heat will cause fouling and increase soap consumption. Light water with a hardness of less than 100 mg/l has a low buffering capacity and may be more corrosive to water pipes.

Sulfate (SO_4^{-2}): Sulfate is naturally present in many minerals and is found in high concentrations in groundwater. Sulfate is one of the anions that has the least toxicity. The presence of sulfate may give rise to a distinct taste in the amount of more than 250 mg/l. Doses of more than 1,200 mg/l of sulfate, along with magnesium, can cause indigestion and dehydration. The WHO standard for sulfate is 250 mg/l.

Chloride (Cl^-): Chloride in drinking water comes from natural sources, domestic wastewater, runoff containing ice-melting salts, and entry of saline water. Excess concentrations of chloride, depending on the alkalinity of water, increase the corrosion rate of metals in the distribution network. As a result, leads to an increase in the concentration of metals in water.

Sodium (Na): Sodium salts are highly soluble in water. Although the concentration of sodium is naturally low in surface water, it may be present in large amounts in some areas and alter the taste of water. Concentrations above 200 mg/l may produce an unacceptable taste for the consumer, and the threshold concentration of sodium flavor depends on the anion and water temperature.

Ammonia (NH_3): Ammonia contains non-ionized NH_3 and ionized NH_4^+. Ammonia in the environment originates from agricultural and industrial processes. The presence of ammonia in water is an indicator of possible bacterial contamination, sewage, and animal waste.

Nitrate and nitrite (NO_3 and NO_2): Nitrate and nitrite are ions that exist as part of the natural nitrogen cycle. In many groundwaters, the increase in nitrate is due to improper disposal of wastewater into absorption wells and the development of agricultural activities. The presence of these compounds can cause methemoglobinemia in infants, and there are concerns about an increased risk of cancer in adults due to N-nitrous compounds. In any case, the amount of nitrate and nitrite should not be judged alone, but the total concentration ratio of each, according to the recommended values, should not exceed one.

Residual free chlorine (Cl_2): To purify and sanitize drinking water, a disinfectant such as chlorine should be used. This substance should be injected into the right amount of water and there should be enough contact time to complete the disinfection operation, and also the rest of the disinfectant and the by-products should not have an adverse health effect. About 5 and 2 milligrams per liter of distilled water in the distilled water, respectively. Most people recognize the taste of chlorine or its by-products in concentrations below 5 milligrams per liter. The taste of chlorine in concentrations between 0.6 and 1 mg/l is recognizable to most people but has no detrimental effect (Eq. 2).

$$\frac{NO_3}{50} + \frac{NO_2}{3} \leqq 1 \qquad \text{(eq. 2)}$$

Iron is one of the most abundant elements in the earth's crust and an essential element in the diet. In refineries that use iron salts for coagulation, the amount of iron in the treated water may increase. Corrosion of steel and cast iron pipes in the distribution network should also be considered. The permissible limit of iron standard in WHO and the national standard of Iran is 3% mg/l.

Manganese (Mn): Manganese is usually present in small amounts in surface water. Excessive manganese ions change the color of water and make it unsuitable for drinking. The concentration of manganese in water without soluble oxygen may reach several milligrams per liter, but the presence of soluble oxygen causes the deposition of manganese salts. Manganese with small amounts is an essential element for the body, but with large amounts and long-term ingestion, it has detrimental effects.

Aluminum (Al): Aluminum is a widely used element with widespread dispersion. In recent years, the widespread use of aluminum salts for coagulation in the treatment process has led to its presence in drinking water, and this is due to the lack of proper operation of the process.

Jar experiment: Determination of the type of coagulant and its appropriate amount is done according to the quality of raw water (especially turbidity and pH) using a flowing device. Determining the optimal amount of coagulant is one of the most important tests in a treatment plant, and in this regard, adjusting the pH value of water in the saturated pH range is very important to prevent sedimentation or corrosion in the drinking water network. Also, the current test indicates the quantitative and qualitative status of the sludge from treatment operation, which will be very useful in regulating the clarification process. An important parameter in this regard is the adhesion coefficient of the sludge [12].

Microbiological Experiments

Understanding the microbiological quality of drinking water is directly related to human and community health. Identification of bacteria, microorganisms, and aquatic organisms that are mainly pathogenic is of particular importance from a health perspective, and drinking water should be free of any pathogenic microorganisms.

Bacterial tests: Bacteria are tiny creatures, some of which are pathogenic and can be transmitted through contaminated water, causing various bacterial diseases. Due to the diversity of pathogenic bacteria in water, control of drinking water is done by identifying and counting the index bacteria (mainly whole films, warm-form films, and fecal streptococci). The counting of heterotrophic bacteria is also important to evaluate the efficiency and effectiveness of the treatment plant [13].

Fatty substancesanalysis: To analyze the fatty substances changes, first the fattysubstance in the samples was extracted from them by a succulent device using organic chloroform solvent and after asphalting in microliter quantities it was injected into a gas chromatography device. The GC device used in this study was a 2010 model made by Winky Company in which courier detection was performed by an FID detector. The 25-meter-long capillary column had an initial temperature of 50 ˚C and a final temperature of 320 ˚C. The temperature rise in the device was set at 5 ˚C per minute. The carrier gas was helium and compressed air and hydrogen gas were used for the FID detector flame. To measure the total organic carbon and amount of total nitrogen, the Rocky Eval 6 device and the Kajdal method were used, respectively.

RESULTS

In the present study, the increase of sewage sludge as soil and air fertilizer during bioremediation along with increasing the growth of heterotrophic bacteria, had a significant effect on fatty substances decomposition so that soil pollution was reduced by 45 to 60%. In the treatment of 100 tons of sewage sludge per hectare (200 grams of sludge in 5 kg of soil), fatty substances analysis was more than treatment of 50 tons of sewage sludge per hectare (100 g of sludge in 5 kg of soil). The results of comparing the average effect of wastewater sludge treatment on fatty substances analysis by Duncan's method at the level of 5% showed a significant difference between control and statistically in treated samples. Therefore, fatty substances decomposition in untreated samples was significantly less than treated samples. Also, the rate of fatty substances decomposition in samples treated with 100 tons of sewage sludge was significantly higher than in samples treated with 50 tons of sewage sludge. Spinora and Dendown (2003) showed that increasing organic matter such as sewage sludge in contaminated soil due to the introduction of large volumes of nutrients accelerates the bioremediation of diesel and decomposition of TPH. The effect of time on fatty substances decomposition at the 5% level was significant, and over time, the rate of fatty substances decomposition decreased. This observation is completely consistent with the results of bacterial growth. The highest bacterial growth was in the fifth week, during which time the activity of bacteria was maximized due to the normal presence of rapidly decomposing alkanes and abundance of mineral nutrients [14]. Over time, the degradable Hydrocarbon compounds remained long-chain, nitrogen-deficient, and bacterial growth and fatty substances degradation decreased. The results of the analysis of variance showed a significant difference between the effects of different sewage sludge treatments on soil C/N ratio. The results of the comparison of the mean by the Duncan method showed a significantly higher C/N ratio in the control compared to the treated samples. However, a significant difference was observed between the two treatments of 50 and 100 in this regard at the level of 5%. In the control sample, mineral deficiency limits the growth of hydrocarbon decomposing bacteria and thus fatty substances decomposition and increases the C/N ratio. As a result, the C/N ratio increased from 6 in the control samples to about 3 in the treated samples. Over time, the C/N ratio dropped significantly, and in the fifth week, it was less than in the tenth week [15].

Analysis of variance of the results obtained from treatment of crude fatty substances contaminated soils with sewage sludge showed that application of sewage sludge treatment had a significant effect on the growth of bacteria in the soil at the level of five percent and there was a significant difference between treated and control samples. During the mineralization process, sewage sludge releases nutrients into the soil solution and increases the growth of fatty substances-decomposing heterotrophic bacteria, fatty substances decomposition and decreases the C/N ratio in the soil by creating suitable nutritional conditions.

In 2000, Ramsey et al. studied the effect of bioremediation on microbial populations in hydrocarbon compounds sediments and found that continuous aeration and increased fertilization significantly stimulated the growth of hydrocarbon-degrading bacteria in the soil. In 2001, Wangstel et al. reported a significant increase in the bacterial population in the bioremediation of fatty substances-contaminated soils.

Kim et al. also reported in 2007 that the effect of fertilization on the biological stimulation of native soil bacteria decreases over time, which was consistent with the results of this study.

Removal of heavy metals using microorganisms

Heavy metals in the field of bioabsorption in three categories: toxic metals (such as Hg, Pb, Zn, Cr, Cd, As, Co, Sn, etc.), precious metals (such as Pd, Pt, Ag, Au, etc.) and radio Nucleoids (such as U, Ra, Am, etc.), which typically weigh more than 5 g/c3, are classified (24, 5). Soft and heavy metal lead is blue-gray and enters the environment through a variety of industries such as ceramics, glass, textiles, batteries, lead bullets for printing letters, etc. Lead enters the human body through air, water and food. The effects and symptoms of lead poisoning appear to be inhibited by its ability to adhere to the protein sulfide group of various enzymes needed to produce heme. Pyrimidine 5-inactivation also inactivates nucleotides (effective in breaking RNA), resulting in the accumulation of ribosomal RNA. Lead prevents the production of dihydroxyvitamin D 1 and 25. The first organs affected by lead are the brain, peripheral nervous system, bone marrow, kidney, and liver. Due to increased consumption and high intrinsic constant, heavy metal pollution has become one of the most important environmental problems today [16]. Conventional methods for removing metal ions from liquid solutions have been studied in detail, such as chemical precipitation, exchange ions, electrochemical treatment, and membrane technologies on activated carbon adsorption, etc. But chemical settling methods and electrochemical operations are ineffective, especially when the concentration of metal ions in the liquid solution is less than 1 to 100 mg/l, and they also produce large amounts of sludge, which is associated with many defects. Ion exchange, membrane technologies, and the process of absorbing activated carbon are extremely expensive, especially when large amounts of heavy metals and water are collected in low concentrations, so they cannot be used on a large scale. Another process is bioabsorption, which uses various natural substances of biological origin, such as bacteria, fungi, yeast, algae, and so on. These natural absorbers can reduce the concentration of heavy ions present in the solution from ppb to ppt. They can effectively separate dissolved metal ions quickly from high-efficiency dilute complex solutions. Therefore, biological adsorption is a good choice for high-volume, low-concentration complex wastewater treatment [17].

CONCLUSION

Wastewater treatment of fat and hydrocarbons products has been a challenge in the chemical industries.industries. The use of treatment systems used in the treatment of municipal wastewater and industries has not been effective in the treatment of wastewater from the chemicalindustries and in addition to the high cost of construction and commissioning and the need for high ground construction, the ability to purify this kind of effluents is not sufficient.

So the effluents of these units have rarely met the existing environmental standards. In general, fatty substances wastewater treatment systems are divided into two stages named pre-treatment and advanced treatment processes. The absorption of heavy metals by microbial cells is the result of biological adsorption mechanisms and biological aggregation.

The term biological adsorption defined the ability of living, inactive, and dead biomass to bind to heavy metals or contaminants in dilute solutions. It is mainly responsible for this property of the cell wall. The term biological accumulation refers to the metabolic uptake by active living cells.

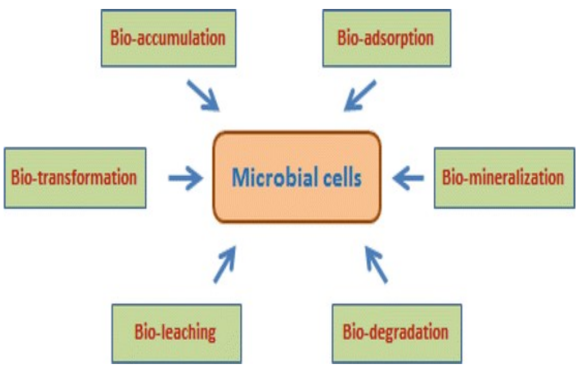

Fig. 2. Bioremediation of Heavy Metals by Microbes

The Biological accumulation of heavy metals in yeast cells has been reported through two stages. The first stage is rapid and non-metabolic and requires binding to the cell wall surface.

In most cases, the ability to bind sucrose and metallic sucrose metals are attributed to electrical charge, which results in weak separation of carboxylic acid groups and other functional groups, such as amino groups that make up the cell wall. The second step in slower and more metabolic-dependent absorption is to store more metal cations than biomass by non-living biomass. However, heavy metals are toxic to microorganisms. Due to their high combined desire to create complexes with the components of membrane, it causes damage to the fragmentation and reduces its performance. Yeast is preferred over other microorganisms for removal of heavy metals due to its survival and growth in heavy metal-contaminated areas, its high metal-cell bonding capacity, and its high rate of intracellular absorption. This study aimed to investigate the removal of heavy metals by microorganisms.

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