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Detoxification of mercury pollutant leached from spent fluorescent lamps using bacterial strains

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ABSTRACT

The spent fluorescent lamps (SFLs) are being classified as a hazardous waste due to having mercury as one of its main components. Mercury is considered the second most toxic heavy metal (arsenic is the first) with harmful effects on animal nervous system as it causes different neurological disorders. In this research, the mercury from phosphor powder was leached, then bioremediated using bacterial strains isolated from Qatari environment. Leaching of mercury was carried out with nitric and hydrochloric acid solutions using two approaches: leaching at ambient conditions and microwave-assisted leaching. The results obtained from this research showed that microwave-assisted leaching method was significantly better in leaching mercury than the acid leaching where the mercury leaching efficiency reached 76.4%. For mercury bio-uptake, twenty bacterial strains (previously isolated and purified from petroleum oil contaminated soils) were sub-cultured on Luria Bertani (LB) plates with mercury chloride to check the bacterial tolerance to mercury. Seven of these twenty strains showed a degree of tolerance to mercury. The bio-uptake capacities of the promising strains were investigated using the mercury leached from the fluorescent lamps. Three of the strains (*Enterobacter helveticus*, *Citrobacter amalonaticus*, and *Cronobacter muytjensii*) showed bio-uptake efficiency ranged from 28.8% to 63.6%.

1. Introduction

Mercury is considered as one of the most toxic metals. Despite this, it is still being used in different applications; one example is fluorescent lamps (Durao et al., 2008; Tunsu et al., 2015). Use of fluorescent lamps had increased in the past years in comparison to incandescent bulbs. This is due to lower energy consumption and longer life expectancy. This translates into lower carbon dioxide emissions, thus a potential reduction of the greenhouse effect (Park et al., 2004). Those fluorescent lamps are composed of glass tube, cathode, anode, phosphor, electronic modules, aluminum end-caps and other constituents e.g. alumina (barrier layer), inert gas and mercury vapor (Harris, 2001).

The mercury elemental form in the fluorescent lamp can be vaporized under high pressure reacting with other components in the lamp (Rhee et al., 2013; Natalia and Gallardo, 2012). The elemental form of mercury is lipid soluble and can pass through both the brain barrier and the placenta which can cause neurological disorders (Nance et al., 2011). Due to the property of being lipid soluble, it then will be absorbed through the skin, gastrointestinal tissues or by inhalation through the respiratory system (Mansur and Fábrega, 2007). It was known that the ionic species of mercury present in fluorescent lamps can form compounds that are more soluble than metallic mercury; thus having a greater impact on the environment (Tunsu et al., 2011, 2015). Raposo et al., 2003 used the thermo-desorption technique to distinguish the mercury species found in the fluorescent lamps. They showed that the mercury species (Hg^0, Hg^+, and Hg^{2+}) happened in the spent fluorescence lamps with the majority of Hg^0 and Hg^+.

Another correlation of exposing to mercury as one of the heavy metals is that it will increase the number of free radicals which will lead to forming lipid peroxidation, damaging the DNA, and protein sulfhydryl depletion (Santos-Gandelman et al., 2014). The amount of mercury in the fluorescent lamps would be different; depending on the manufacturing company, and the state of the lamp; for how many years it was used (Gallardo and Rey-Raap, 2012). Different studies were showed that a normal fluorescent lamp contains about 30–40 mg of mercury (dos Santos et al., 2010) and the average concentration of mercury is roughly 7.2 mg. dos Santos et al. (2010) analyzed several fluorescent lamps and found that the mercury masses per lamp in 15 lamps were in the range from 1.6 to 27 mg and they were above the limit allowed (5 mg per compact lamp) by the European community in six samples.

Owing to the moderately small amounts of mercury contained in fluorescent lamps, the fluorescent lamps are considered a

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hazardous waste (Tunsu et al., 2011; Wagner-Dobler, 2013) and their disposal in municipal solid waste (MSW) management facilities should be prohibited. The environmental impact due to mercury contents in fluorescent lamps can be diminished by reducing the mercury contents inside the lamps with innovative methodology and increasing the lamp recycling rate.

Mercury must be treated before disposing it by first separating the phosphor powder from the lamps, then leaching the mercury from the phosphor powder which is much harder due to the strong interactions between them (Tunsu et al., 2011). The recycling usually focus on the phosphor powder because 80% of mercury reacts with it (Rhee et al., 2013). For their economic and environmental efficiency, leaching of mercury was carried out with thermal desorption, nitric and hydrochloric acid solutions using two approaches: leaching at ambient conditions and microwave-assisted leaching (Jang et al., 2005; dos Santos et al., 2010).

Environmental remediation of mercury is one of the key tasks for sustainable development. Scientists are using different methodologies to eliminate the level of mercury in the wastes. Several physicochemical techniques for elimination of mercury have been implemented in order to accomplish the required environmental guidelines such as adsorption, chemical, precipitation, filtration, thermal desorption and ion exchange (Jang et al., 2005; dos Santos et al., 2010; Natalia and Gallardo, 2012). However, some of these techniques have shortcoming such as costly, less effective and sometime produce harmful by-products such as chemical sludge which in turn creates disposal problems (Barakat, 2011; Wagner-Dobler, 2013). Therefore, new technologies to eliminate mercury are of importance. As an alternative to traditional physical and chemical decontamination techniques, hydrogen sulfide producing yeasts can be used in bioremediation. In this case elemental mercury can be converted to less volatile and more stable species, e.g. mercury sulfide, according to the equation ($Hg + S \rightarrow HgS$) (Latif and Amin, 2011; Cho et al., 2013; López et al., 2015). Therefore, microbial bioremediation is an interesting methodology for mercury remediation and it appears as an attractive methodology owing to its low cost, simple, and environmental friendly (Wagner-Dobler, 2013; Chaturabul et al., 2015). Green biotechnology may have a safer way to reduce the toxicity of mercury. Certain species of bacteria, (either gram positive or gram negative) are known to have operons which control a gene known as "mer operon". This operon is on the plasmid of the bacteria that allows the bacteria to change the form of mercury from toxic to more stable form (Osborn et al., 1997). This operon can be functional for seven different genes (merA, merB, merC, merD, merF, merT and merP) (Das and Dash, 2013). The Mer genes are located on the plasmid, transposons and integrons (Dea et al., 2014). Transposons is part of the DNA which can change its location, also known as jumping DNA, it is being carried on the plasmid itself. There are 29 known mercury resistance transposons (Mindlin et al., 2001). Three mercury resistance transposons are well studied which are Tn*21*, Tn*5053*, and Tn*501* (Reniero et al., 1998). Another mercury resistance transposon is Tn502 and Tn512 taken from *Pseudomonas* strain (Petrovski et al., 2011). Moreover the expression of these genes is positively induced by mercury and also negatively regulated by the transcriptional factors which are regulated in the cytosol. Since the Mer operon is positively induced by the mercury, it must reach the cytosol to start the transcription which is carried out by (MerP + MerT) (Reniero et al., 1998). MerC and MerF code for the membrane proteins and the other parts of the Mer operon are coding for auto regulated genes as MerR and MerD, however MerG is the only one that codes for phenylmercury resistance (Reniero et al., 1998). Depending on the gene varieties, the bacteria can be classified as broad-spectrum resistant (with MerB) or narrow-spectrum resistant (Dea et al., 2014). Also these varieties can be different depending on the bacteria if it is gram positive or gram negative, with gram negative possesses more resistance to mercury (Abou-Shanab et al., 2007). Previous studies highlight a range of microorganisms that have mer operon and able to reduce Hg^{2+} to elemental Hg (Yu et al., 2014). Some bacteria have resistance to mercury by synthesizing thiol compounds which then react with the mercury and reduce its effect (Wagner-Dobler, 2013; Dea et al., 2014). Another mechanism is having a permeability barrier where the mercury access to the cell is reduced (Das and Dash, 2013). In sight of the ill-effects of the SFLs wastes to both environment and health, Qatar has encouraged the need to address the problems and challenges posed by hazardous waste. Leaching and bio-uptake of mercury in phosphors from SFLs could be a promising approach to treat and dispose them in a safe way when the waste of the spent fluorescent lamps effectively collected. Accordingly, the need for environmentally comprehensive management of waste of the SFLs has become more and more urgent in Qatar. Therefore, the objectives of this study are: (i) to leach the maximum amount of mercury from the SFLs (phosphor powder) using acid and microwave-assisted leaching, (ii) to treat the leached mercury using indigenous bacterial strains isolated from Qatari soil polluted with petroleum to reduce the toxicity of the mercury.

2. Methodology

2.1. Acid leaching

T12 and T8 spent fluorescent lamps (34 lamps) were collected from different brands as Philips and Chiyoda companies. Then the lamps were broken near the aluminum cap and the interior surface of the lamps was scratched using a spatula in order to collect the maximum amount of the powder. The total amount of the phosphor powder was 170.5 g. T12 and T8 spent fluorescent lamps contained approximately 4–6 g of white powder. Eight different ratios of hydrochloric acid (37% v/v HCl) and nitric acid (68% v/v HNO_3) (Table 1) were prepared and the solution was diluted using distilled water up to 100 ml and each treatment was repeated three times. A specific weight of the phosphor powder (0.25 g) was added into the prepared acid solution (S/L = 2.5 g/l), and then the samples were placed on a shaker overnight at 25 °C and 130 RPM. After that, the samples were filtered using syringe-filter of a 0.2 μm along with Millipore filtration unit. Finally, the samples were placed in the refrigerator at −4 °C for mercury analysis using the cold vapor atomic absorption spectrometry analysis (CVAAS).

2.2. Microwave-assisted leaching

Microwave-assisted leaching experiments were prepared with three different ratios of hydrochloric acid (37% v/v HCl) and nitric acid (68% v/v HNO_3) (Table 1) along with 0.5 ml of hydrogen peroxide (10% v/v H_2O_2) (as oxidizing agent) and then a specific weight of the collected phosphor powder (0.25 g) (S/L = 2.5 g/l) was added into the solution. Later on, the samples were diluted up to 50 ml of distilled water, and were placed in the microwave (Panasonic "Inverter") in different powers and times (Table 1). The plastic caps of the bottles were taken off to prevent overheating and boiling of the samples which might create an accident. The thirty-six samples were taken out from the microwave and left to cool down. After couple of hours, they were filtered out using a syringe-filter of 0.2 μm. The samples were then stored in a refrigerator at 4 °C until the CVAAS analysis.

2.3. CVAAS analysis

To measure the amount of mercury which was leached by the two above-mentioned methods, cold vapor atomic absorption