Chemosphere 207 (2018) 753–763

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

A statistical approach of zinc remediation using acidophilic bacterium via an integrated approach of bioleaching enhanced electrokinetic remediation (BEER) technology

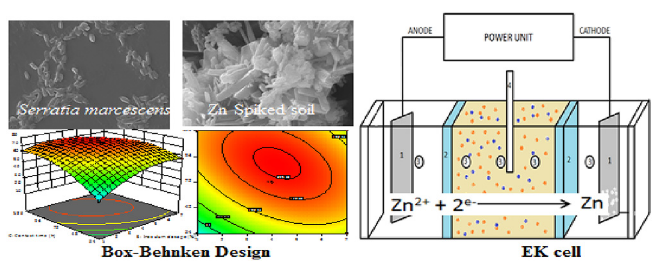
Adikesavan Selvi**, Rajasekar Aruliah*

Environmental Molecular Microbiology Research Laboratory, Department of Biotechnology, Thiruvalluvar University, Serkkadu, Vellore, 632115, Tamilnadu, India

HIGHLIGHTS

- An acidophilic bacterium was isolated from tannery effluent contaminated sludge.
- Bioleaching and Electrokinetic process parameters were optimised using a statistical model.
- An integrated method of Bioleaching Enhanced Electrokinetic remediation was demonstrated for zinc removal in real soil.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 March 2018
Received in revised form
17 May 2018
Accepted 24 May 2018
Available online 25 May 2018

Handling Editor: X. Cao

Keywords:
Acidophilic
Bioleaching
Electrokinetic
Response surface methodology
Zinc

ABSTRACT

The aim of the present study was to isolate an indigenous acidophilic bacterium from tannery effluent contaminated sludge (TECS) sample and evaluate its potentiality towards the removal of zinc using an integrated approach of bioleaching enhanced electrokinetic remediation (BEER) technology in zinc spiked soil at an initial concentration of 1000 mg/kg. The isolated acidophilic bacterium was characterized by biochemical and 16S rRNA molecular identification and was named as *Serratia marcescens* SMAR1 bearing an accession no. MG742410 in NCBI database. The effect of pH and inoculum dosage of SMAR 1 strain showed an optimal growth at pH 5.0 and 4% (v/v) respectively. Based on these experimental data, a statistical analysis was done using Design Expert computer software, v11 to study the interaction between the process parameters with respect to zinc reduction as an output response. Electrokinetic experiments were conducted in a customised EK cell under optimised process conditions, employing titanium electrodes. Experiments for zinc removal were demonstrated for bioleaching, electrokinetic (EK) and BEER technology. On comparing, the integrated process was found to evidence as an excellent metal remediation option with a maximum zinc removal of 93.08% in 72 h than plain bioleaching (72.86%) and EK (56.67%) in 96 h. This is the first report of zinc removal in a short period of time using *Serratia marcescens*. It is therefore concluded that the BEER approach can be regarded as an effective technology in cleaning up the metal contaminated environment with an easy recovery and reuse option within short period of time.

1. Introduction

Heavy metals constitute a group of highly persistent

* Corresponding author.
** Corresponding author.
E-mail addresses: seljeev@gmail.com (A. Selvi), rajasekargood@gmail.com, rajasekargood@tvu.edu.in (R. Aruliah).

https://doi.org/10.1016/j.chemosphere.2018.05.144

environmental contaminants, due to which their effect on biotic forms are highly extreme. Their nature of resistant to degradation by natural means has led to bioaccumulation and biomagnification of heavy metals in the food chain (Järup, 2003; Rzymski et al., 2014). Though these metals exists as natural elements of the earth's crust, their existence in the soil could be greatly contributed to various sources of anthropogenic activities like, improper disposal of electronic waste, industrial discharges, improper domestic waste management practices, unorganised dumping, use of fertilizers, transportation, etc. (Herawati et al., 2000; Goyer, 2001; He et al., 2005; Szyczewski et al., 2009). As a result of these, a drastic alteration in the biochemical balance and geochemical cycles have been reported (Bradl, 2002).

In general, few of the heavy metals namely, zinc (Zn), copper (Cu), manganese (Mn), nickel (Ni) etc., are grouped as essential micronutrients, that are needed at very low concentration for certain biological metabolism in living organisms (WHO/FAO/IAEA, 1996; Wang et al., 2014). On the other hand, heavy metals like, mercury (Hg), lead (Pb), cadmium (Cd) and Arsenic (As) etc., don't have any specific metabolic role and hence are categorised as obligatory toxic (Rzymski et al., 2015). However, both the group of heavy metal's uptake could be highly toxic at higher concentrations causing serious health effects like tubular damage in kidneys, lung damage (Mandel et al., 1995; Hotz et al., 1999), coronary heart disease (Salonen et al., 1995), lung cancer, stomach cancer and gliomas (IARC, 1993; Steenland and Boffetta, 2000; WHO, 2001) brain damage, psychological and neurological symptoms, such as shivering, personality changes, anxiety, restlessness, sleep disturbances and depression (Järup, 2003).

Over the past, various heavy metal removal/recovery methods like, chlorination (Fraissler et al., 2009), chemical extraction (Marinos et al., 2007), electrokinetics (Violetta and Sergio, 2009), ion exchange (De Villiers et al., 1995), membrane separation (Chaudry et al., 1997), and bioleaching methods (Pathak et al., 2009) from soil/sludge have been reported by many researchers. Owing to advantages such as, cost-effectiveness, efficiency and less energy consumption, biological methods are much preferred than any other methods (Fang et al., 2014; Chi and Gao, 2015). Bioleaching is one such effective approach that is commonly employed to recover/remove the heavy metals from soil/sludge. In this method, the metals are dissolved under the acidic condition by the action of acidophilic microorganisms (Pathak et al., 2009; Govarthanan et al., 2014). Unfortunately, factors like, microbial acclimatization time and biodegradable efficiency of the indigenous microorganisms greatly limits its application, if the process is employed alone (Mrozik and Piotrowska-Seget, 2010). Likewise, electrokinetic remediation (ER) method is also equally preferred as biological methods because of its simple operation, effectiveness, and no subsequent pollution for the removal of heavy metals from soil/sludge (Zhou et al., 2004; Deng et al., 2009; Violetta and Sergio, 2009; Ma et al., 2010). Similar to bioleaching, ER method has also certain restrictions like, bioavailability of the heavy metal and mass transfer between the electrode and pollutants (Lohner and Tiehm, 2009; Simoni et al., 2001).

But these restrictions can be overcome by combining bioleaching with electrokinetic (EK) remediation method, as this combination is believed to promote increased bioavailability of the pollutants, enhancement in biodegradation efficiency by generating oxidization and reduction zones, releasing of soil/sediment bound pollutant, improved nutrient transport, improved performance, and availability of terminal electron acceptors (Maini et al., 2000; Luo et al., 2005; Wick, 2009; Kim et al., 2010; Peng et al., 2011). In addition, the bioleaching process combined with electrokinetic technology is considered advantageous with an extended function of recycling the metals for other applications.

Since we are integrating two different approaches, the possible outcomes of the experiment are evaluated using RSM software by optimizing different variables (pH, inoculum dosage and contact time) chosen for the present study. This is done to reduce the time and the number of experimental trials by calculating the number of possibilities of variables and their interactions. Out of other models, Box-Behnken design (BBD) used in our study is considered to be one of the superior statistical designs which mathematically compute the significance of many variables in less number of experiments (Ojha and Das, 2018).

Therefore, the present study is carried out to demonstrate the feasibility of heavy metal removal using an integrated approach of bioleaching and enhanced electrokinetic remediation (BEER) technology at lab scale, under optimised operative conditions on real soil spiked with zinc. In this study, we have chosen zinc (Zn) as a model pollutant because of its increased soil concentration due to numerous anthropogenic inputs, globally (Zhang et al., 2012; Cruz et al., 2014; Moreira et al., 2016; Lu et al., 2017). Moreover, Zn is considered to be one of the most mobile and potentially toxic heavy metals present in the environment (Mishra et al., 2008; Gondek and Mierzwa-Hersztek, 2016). Their non-volatilising nature and partial leaching were reported to cause acute and chronic effects in both aquatic and terrestrial biota (Chen et al., 2010; Milosavljević et al., 2011). Presence of Zn in soils, more than 0.6 mg/kg was reported to interfere with plant metabolism in absorbing other essential metals, like manganese and iron (WHO, 1996; Cameron, 1992). Hence, an integrated attempt demonstrated in our present study is expected to remediate polluted territory to pristine state.

2. Materials and methods

2.1. Chemicals and reagents

Luria-Bertani (LB) agar, Tris Hcl, EDTA, Glucose, and Zinc sulphate were procured from Himedia Laboratories, India, Pvt. Ltd. Sulphuric acid (95–98%) and nitric acid (>90%) were purchased from Sigma-Aldrich, India, Pvt. Ltd. All the chemicals and acid solutions used in this study were of analytical grade and used without further purification.

2.2. Sample collection

The tannery effluent contaminated sludge (TECS) was collected from a tannery industry located at Peranambut (12.9447° N, 78.8708° E), Vellore District, Tamilnadu, India. A sample size of 10 kg was collected in a clean plastic bucket provided with lid, using a clean mini-shovel. The samples were kept at 37 °C until we reached laboratory from the collection site. The samples were stored at 4 °C, until isolation.

2.3. Enrichment of acidophilic microorganisms

The enrichment procedure was carried out following the protocol by Xiang et al. (1998) with slight modifications. Briefly, 50 g of the collected soil was added to 100 ml of distilled water and incubated at 30 °C in a shaker incubator at 150 rpm for 7 days. 3 g/L of glucose was added as a sole carbon source. At the end of incubation, 10 ml of the soil suspension from the above culture flasks (test flasks) was transferred to five separate Erlenmeyer flasks containing 100 ml of sterile distilled water. The pH was adjusted to 2.0–6.0 using 1 N H_2SO_4. Sterile Erlenmeyer flasks containing sterile distilled water adjusted to the pH range of 2.0–6.0, served as a control. The set up was incubated at 30 °C in a shaker incubator at 150 rpm for 3 days.