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Bioleaching of gallium from gallium arsenide by *Cellulosimicrobium funkei* and its application to semiconductor/electronic wastes

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ABSTRACT

The aim of this work was to screen and characterize heterotrophic bacteria for gallium arsenide (GaAs) leaching. Ga in the form of GaAs has been extensively used as a semiconductor substrate material. The advantage of using microbes for gallium recovery is the fact that this method is a safer, environment-friendly and includes energy-saving processes, which can leach metals at relatively low concentrations. Eight bacterial isolates were isolated from cadmium-, and arsenic-contaminated soil in the presence of GaAs. Pad I and NKS III showed the highest efficiency in Ga leaching at approximately 63–81% after 15 and 30 days, respectively. The analysis of 16S rDNA sequences indicated that strain Pad I was close to strain NKS III; it was characterized and identified as *Cellulosimicrobium funkei* (*C. funkei*). Further investigation revealed that the ability of Ga leaching from GaAs by this bacterium involved amino acids. This process occurred in a weak base pH range. The results show a potential application of *C. funkei* to leach Ga from semiconductors or electronic wastes. The in-depth mechanism of bacterial leaching of Ga from GaAs was also investigated.

1. Introduction

Nowadays, electronic waste (E-waste) is among the most rapidly growing problems in the world. It comprises discarded electronic appliances, such as computers, television sets and cell phones, which are increasing at a high rate because of their short lifespan (Robinson, 2009) and human lifestyle that follows new modern trends. E-waste contains both valuable and hazardous materials (e.g. lead, mercury, arsenic, cadmium, selenium and hexavalent chromium); (Pant et al., 2012) that require management to avoid environmental contamination and harmful effects on human health (Robinson, 2009). Several methods were used for management of E-waste; they include disposal into landfills, reusing, remanufacturing and recycling (Cui and Zhang, 2008). Landfills might be problematic due to toxic chemicals leaching into the surrounding environment, particularly to soil and water, which may later be harmful for people who live close to landfills. Reusing is the priority for the management of E-waste since the lifespan of usable equipment can be extended on a secondary

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market, resulting in a reduction in the volume of treated waste stream. Remanufacturing is a production-batch process where used products or cores are disassembled, cleaned, repaired or refurbished, and tested to produce new or like-new equipment (Williams and Shu, 2001). Recycling means reprocessing in a production of the waste materials for the original purpose such as cell phone, television or for other purposes. Recycling of E-waste involves disassembly and/or destruction of used equipment in order to recover materials (Cui and Zhang, 2008). Currently, recycling of E-waste can be broadly divided into three major stages. The first stage which is disassembly or selective disassembly, aims at singling out hazardous or valuable components; it is an indispensable process. The second stage is upgrading, using mechanical/physical processing and/or metallurgical processing to upgrade the contents of desirable materials, i.e. preparing materials for refining process. The last stage is refining, which is recovering materials and returning them to their life cycle (Cui and Forssberg, 2003).

Recovering valuable metals from E-waste is an important subject; it normally involves pyrometallurgical (thermal), hydrometallurgical (chemical) and biometallurgical (biological) processes. For instance, pyrometallurgy employs heat treatment, such as roasting and smelting while hydrometallurgical approach employs leaching metals mainly by acid washing. These two processes are good techniques; however, they still have their own limitations. They are very rapid and efficient but have their own environmental impact due to a release of toxic gases such as dioxins and furans, and discharge of high volume acid wastewater (Cui and Zhang, 2008). Biological leaching, on the other hand, is a comparatively cost-effective technique, that is based on the natural ability of microbes to transform solid metallic compounds to their soluble and extractable forms.

Gallium (Ga) is a valuable metal of great interest. Gallium as gallium arsenide (GaAs) has been extensively used as a substrate material for semiconductors. GaAs-based semiconductor devices are widely used in the world. They are found in commercial goods and have numerous military applications including radio frequency chipsets in cell phones, night vision telescopes, laser diodes in high frequency communication systems, light-emitting diodes (LED) for displays and photovoltaic applications in satellites, etc. (Torrance et al., 2010). Due to high demand for advanced communication systems and wireless semiconductor devices, the availability of Ga/GaAs has become immensely important.

Thus, the aim of this research is to screen and identify microorganisms that can extract or leach Ga from GaAs and be applied in leaching of semiconductors and electronic waste. The isolate *Cellulosimicrobium funkei*; Pad I (*C. funkei*) used in *in situ* leaching compared to growth supernatant was studied. The mechanism involved in Ga leaching by *C. funkei* was also investigated.

2. Materials and methods

2.1. Materials

2.1.1. Gallium arsenide (GaAs)

GaAs was purchased from Sigma–Aldrich (St. Louis, USA). GaAs was ground with standard lab mortar and pestle, sieved through standard 400 mesh screen (particle size smaller than 38 μm) and stored in a desiccator after drying in an oven overnight at 60 °C.

2.1.2. Thin film GaAs solar cells waste

Thin film GaAs solar cells waste was obtained from Department of Physics, Faculty of Science, Chulalongkorn University, Bangkok, Thailand. It was prepared in the same way as GaAs and used in Section 3.5.

2.1.3. Heavy metal-contaminated soil samples

Heavy metal-contaminated soils which were used to isolate GaAs-leaching bacteria were cadmium-contaminated soil from Phra Tad Padaeng Sub-district, Mae Sot District in Tak Province, Thailand; arsenic-contaminated soil from Ronpiboon Sub-district, Ronpiboon District in Nakhon Sri Thammarat Province.

2.2. Screening and isolating gallium arsenide (GaAs)-leaching bacteria

1% (w/v) of each heavy metal contaminated-soil and non-sterile GaAs was used to prepare the initial stock of mixed bacterial culture in an enrichment medium (Luria Bertani (LB) medium: 1% tryptone, 0.5% yeast extract, 1% sodium chloride) at pH 7, 150 rpm and 30 °C overnight. Thereafter, 4% (v/v) of 1×10^8 CFU mL^{-1} of each culture stock were used to screen GaAs-leaching bacteria in 25 mL of LB medium containing 5 mg of sterile GaAs under neutral conditions (pH 7), at 150 rpm and 30 °C for 30 days. The population of mixed microbes which showed the highest efficiency for Ga leaching was isolated by spread plate technique and pure isolates were obtained by repeated streak plate method on agar LB medium and investigated the ability to leach Ga from GaAs under the same conditions. The strain which showed the highest efficiency of Ga leaching was selected for further study.

2.3. Gallium concentration analysis

At the end of bioleaching experiment, the sample solutions were topped up to the same level (25 mL) with sterile distilled water as required to compensate for water loss due to evaporation. The final sample solution was collected, centrifuged at 4500 rpm for 10 min to separate medium solution from the bacterial cells, and then measured the samples by inductively coupled plasma spectroscopy (ICP). The efficiency of Ga leaching from GaAs was calculated according to the following Eq. (1):

$$\%\,\text{Ga leaching} = \frac{C_L \times 100}{C_I} \quad (1)$$

where C_I is the initial Ga concentration in the solution = 86.25 mg L^{-1} (total Ga was leached from 5 mg of GaAs in 25 mL of LB medium) and C_L is Ga concentration (mg L^{-1}) in the solution after leaching process.

2.4. Molecular characterization

The isolated GaAs-leaching bacteria which showed high ability to leach gallium were chosen for further identification by 16S rDNA sequence analysis. The steps for the characterization included isolation of genomic DNA followed by PCR amplification of universal 16S rDNA gene and using 8F (5′-AGAGTTTGATCCTGGCTCAG-3′) and 1492R (5′-GGTTACCTTGTTACGACTT-3′) primers. The amplified fragments were cloned using a pGEM-T Easy Vector Cloning Kit (Promega), as described in the manufacturer's instructions,