Science of the Total Environment 639 (2018) 497–503

Contents lists available at ScienceDirect

Science of the Total Environment

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

Leaching of vanadium from waste V_2O_5-WO_3/TiO_2 catalyst catalyzed by functional microorganisms

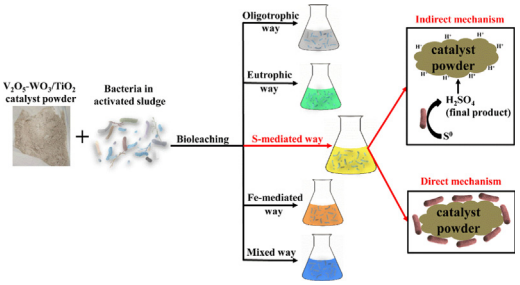
Shuhua Wang [a,b], Yaling Xie [c], Weifu Yan [a,b], Xuee Wu [c], Chin-Tsan Wang [d], Feng Zhao [a,*]

[a] CAS Key Laboratory of Urban Pollutant Conversion, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China
[c] College of Chemistry and Chemical Engineering, Xiamen University, Xiamen 361021, China
[d] Department of Mechanical and Electro-Mechanical Engineering, National I-Lan University, I-Lan 260, Taiwan

HIGHLIGHTS

- Bioleaching of waste V_2O_5-WO_3/TiO_2 catalyst was performed using five methods.
- The S-mediated way was demonstrated as a promising method to extract vanadium.
- Direct and indirect mechanisms worked to leach vanadium effectively.
- The direct vanadium bioleaching mechanism of the S-mediated way was first reported.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 April 2018
Received in revised form 13 May 2018
Accepted 14 May 2018
Available online xxxx

Editor: Jay Gan

Keywords:
Solid wastes
Selective catalytic reduction catalyst
V_2O_5-WO_3/TiO_2
Bioleaching
Acidithiobacillus

ABSTRACT

Solid wastes are currently produced in large amounts. Although bioleaching of metals from solid wastes is an economical and sustainable technology, it has seldom been used to recycle metals from abandoned catalyst. In this study, the bioleaching of vanadium from V_2O_5-WO_3/TiO_2 catalyst were comprehensively investigated through five methods: Oligotrophic way, Eutrophic way, S-mediated way, Fe-mediated way and Mixed way of S-mediated and Fe-mediated. The observed vanadium bioleaching effectiveness of the assayed methods was follows: S-mediated > Mixed > Oligotrophic > Eutrophic > Fe-mediated, which yielded the maximum bioleaching efficiencies of approximately 90%, 35%, 33%, 20% and 7%, respectively. The microbial community analysis suggested that the predominant genera *Acidithiobacillus* and *Sulfobacillus* from the S-mediated bioleaching way effectively catalyzed the vanadium leaching, which could have occurred through the indirect mechanism from the microbial oxidation of S^0. In addition, the direct mechanism, involving direct electron transfer between the catalyst and the microorganisms that attached to the catalyst surface, should also help the vanadium to be leached more effectively. Therefore, this work provides guidance for future research and practical application on the treatment of waste V_2O_5-WO_3/TiO_2 catalyst.

1. Introduction

Selective catalytic reduction catalyst (SCR catalyst) has been extensively used in fossil fuel combustion plants (Kim et al., 2015). V_2O_5-WO_3/TiO_2 is a traditional commercial SCR catalyst, containing approximately 1–5% V_2O_5 and 10% WO_3 (Castellino et al., 2008; Kim et al., 2015; Kompio et al., 2012). Both vanadium and tungsten are toxic to human and environmental health but play a pivotal role in industrial applications, such as in the metallurgy and aerospace industries (Amiri et al., 2011; Li et al., 2014; Nikiforova et al., 2016; Zhang et al., 2007). However, this catalyst has a limited life with 2–3 years due to

* Corresponding author.
E-mail address: fzhao@iue.ac.cn (F. Zhao).

https://doi.org/10.1016/j.scitotenv.2018.05.168
0048-9697/

inactivation (Benson et al., 2005; Li et al., 2016; Madia et al., 2002; Nova et al., 2001). In China, it is estimated that 38,000 t/y of catalyst becomes solid wastes (Li et al., 2014). Therefore, extracting metals from waste catalyst would alleviate the national demand for metals and mitigate damage to the environment.

Hydrometallurgy, pyrometallurgy and bioleaching are the general methods used for metal recovery from solid wastes. Both hydrometallurgical and pyrometallurgical processes can contribute to metal mixtures being rapidly and efficiently obtained, resulting from high acid or alkali and high temperature conditions, respectively (Aarabi-Karasgani et al., 2010; Havlik et al., 2010; Huo et al., 2015). However, these two processes produce environmental pollutants, such as dioxins, furans and highly acidic wastewater (Gu et al., 2017). Bioleaching of metals from solid wastes is a promising and economical technology with low-cost and environment-friendly benefits (Fonti et al., 2017). Bioleaching has been widely applied to solubilize metals from solid wastes such as swine manure and electronic wastes (Wei et al., 2018; Xiang et al., 2010), but few studies have concentrated on waste catalysts. Unlike printed circuit boards (PCBs, the classic electronic waste), in which the metals are primarily in the form of elementary substances (Hong and Valix, 2014; Li et al., 2015), the elements in the V_2O_5-WO_3/TiO_2 catalyst exist as oxides. In addition, TiO_2 is anatase polymorphic, while monomeric vanadyls and wolframyls and polymeric W_WO_Y are observed for V_2O_5 and WO_3 (Alemany et al., 1995). Thus, there are markedly different physicochemical structures and properties between PCBs and V_2O_5-WO_3/TiO_2 catalyst, which may lead to different bioleaching mechanisms.

Therefore, in this study, we investigated five different bioleaching pathways to extract vanadium and tungsten from waste V_2O_5-WO_3/TiO_2 catalyst, including a simple carbon-mediated pathway (Oligotrophic way), rich carbon-mediated pathway (Eutrophic way), reduced sulfur-mediated pathway (S-mediated way), iron-mediated pathway (Fe-mediated way) and a mixed of sulfur- and iron-mediated pathway (Mixed way) (Brandl et al., 2001; Chi et al., 2011; Choi et al., 2004; Wang et al., 2009; Wang et al., 2016). The most important distinction among these five methods was the diverse lixiviant produced by different functional microorganism communities that grew on different electron donors. To understand the bioleaching mechanism, the microbial community structure was investigated using amplification sequencing. Accordingly, the major objective of this study was to fully explore the feasibility of using different bioleaching methods for V_2O_5-WO_3/TiO_2 catalyst recycling to identify an economical and eco-friendly method.

2. Materials and methods

2.1. Waste V_2O_5-WO_3/TiO_2 catalyst

The industrial waste catalysts used in the experiment were kindly provided by Prof. Jinsheng Chen (Institute of Urban Environment, Chinese Academy of Sciences, China). After peeling off the flat, non-catalytic outer layer, the catalysts were mashed by a pestle and then shattered using a multi-function disintegrator (Xiaobao, China) at a speed of 31,000 rpm for 10 min. The powders were sieved and particle sizes below 80-mesh were collected. The final catalyst powders were dried in a vacuum oven at 75 °C to a constant weight before they were used in experiments. The metal determination of spent catalysts was performed according to a previously described method (Wang et al., 2016). The vanadium and tungsten contents are shown in Table S1 (Supporting information). The titanium was not detected, as it barely solubilized.

2.2. Microorganisms and culture medium

Experimental microbial communities were enriched from aerobic activated sludge (supplied by the Jimei sewage treatment plant in Xiamen, Fujian province, China). The bacteria were cultivated in M9, LB, Starkey, 9K and Fe+S media, containing glucose (Oligotrophic way), peptone (Eutrophic way), S^0 (S-mediated way), Fe^{2+} (Fe-mediated way) and Fe^{2+}+S^0 (Mixed way) as energy sources. The components of M9 medium (g/L): NaCl, 0.5; NH_4Cl, 1; K_2HPO_4, 3; Na_2HPO_4, 6. The medium pH was approximately 7.0 without adjustment and was sterilized under 121 °C for 20 min. Then 1 mL each of 1 M $MgSO_4 \cdot 7H_2O$ and 0.1 M $CaCl_2 \cdot 2H_2O$ which have been autoclaved at 121 °C for 20 min were added to the solution in a sterile environment. Finally, the glucose was added to the solution to a final concentration of 4 g/L after autoclaved at 115 °C for 15 min (Reeve et al., 1984).The LB medium contained the following (g/L): Peptone, 10; Yeast extract, 5; NaCl, 5. After the pH was adjusted to 7.2 using NaOH, the medium was autoclaved at 121 °C for 20 min. Filter sterilized glycine solution was added to the solution to a final concentration of 5 g/L in a sterile environment before inoculation (Brandl et al., 2008). The Starkey medium was composed of (g/L): $MgSO_4 \cdot 7H_2O$, 0.5; $(NH_4)_2SO_4$, 0.4; $CaCl_2 \cdot 2H_2O$, 0.25; K_2HPO_4, 3; $FeSO_4 \cdot 7H_2O$, 0.01; Yeast extract, 0.5. The pH was adjusted to 2.0 using sulfuric acid and was autoclaved at 121 °C for 20 min. 10 g/L sulfur was added to the medium in the clean bench after intermittent sterilization (Findley et al., 1974; Wang et al., 2009). The 9 K medium contained (g/L): KCl, 0.1; $(NH_4)_2SO_4$, 3; $MgSO_4 \cdot 7H_2O$, 0.5; K_2HPO_4, 0.5; $Ca(NO_3)_2$, 0.01; Yeast extract, 0.5. Before autoclaved at 121 °C for 20 min, adjusting the medium pH to 2.0 using sulfuric acid. Filter sterilized $FeSO_4 \cdot 7H_2O$ solution was added to the solution to a final content of 44.7 g/L in the clean bench before inoculation (Wang et al., 2009). The Fe+S medium contained the following (g/L): KCl, 0.1; $(NH_4)_2SO_4$, 2; $MgSO_4 \cdot 7H_2O$, 0.25; K_2HPO_4, 0.1; Yeast extract, 0.5; Sulfur, 5; $FeSO_4 \cdot 7H_2O$, 22.35 (Wang et al., 2009). The medium pH was 2.0, and the sterilization method was the same to Starkey and 9K.

To obtain the functional microbial communities, 10 mL of sludge and a specific amount of catalyst powders were added into 250 mL flasks containing 90 mL of M9, LB, Starky, 9K and Fe+S media, respectively. The flasks were incubated at 30 °C in a shaking incubator at 150 rpm for 7 days, after which 10 mL of culture was transferred to fresh M9, LB, Starky, 9K and Fe+S media with different amounts of catalyst powder.

2.3. Bioleaching experiments

To identify the most economical and eco-friendly bioleaching method, the experiments were conducted to assess the above-mentioned five bioleaching pathways. After two months of acclimation, five types of microbial communities were used in the bioleaching experiments, and the initial content of catalyst was 10 g/L. Abiotic experiments were conducted under the same conditions. To investigate the influence of different initial amounts of catalyst powder on bioleaching efficiency, a series of flasks were arranged with various amounts of catalyst powders. Each experiment was carried out in triplicate, and the average data were reported.

2.4. Analytical methods

During the experiment, portions of the bioleaching solutions were periodically taken to test the pH and the concentrations of soluble V, W and Fe^{2+}. Each sample was firstly filtered through a 0.22 μm membrane to remove cell debris and precipitates prior to measurements. The pH was measured using a pH meter (UB-7, Denver, USA) and the concentration of Fe^{2+} was determined using a previously described ferrozine assay (Panda et al., 2017). Next, a certain amount of filtrate was diluted to an appropriate concentration range with 2% HNO_3, after which the concentration of soluble V and W were determined by inductively coupled plasma-optical emission spectrometry (Optima 7000DV, PerkinElmer, USA).