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Selective Mineralization and Recovery of Au(III) from multi-ionic aqueous systems by Microorganisms

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Abstract:

The recovery of precious metals is a project with both economic and environmental significance. In this paper, it presents how to use bacterial mineralization to selectively recover gold from multi-ionic aqueous systems. The *Bacillus licheniformis* FZUL-63, separated from a landscape lake in FuZhou University, was shown to selectively mineralize and precipitate gold from coexisting ions in aqueous solution. The removal of Au(III) was almost happened in first hour, and FTIR data show that the amino, carboxyl and phosphate groups on the surface of the bacteria are related to the adsorption of gold ions. XPS results implied that Au(III) ions are reduced to monovalent, and then the Au(I) was adsorbed on the bacterial surface at the beginning stage(first hour). XRD results showed the gold biomineralization began about 10 hours after the interaction between Au(III) ions and bacteria. The Au(III) mineralization has been rarely influenced by other co-existing metal ions. TEM analysis shows the gold nanoparticles are polyhedral structure with a particle size of ~20 nm. The *Bacillus licheniformis* FZUL-63 could selectively mineralize and recover 478 mg/g(dry biomass) gold from aqua regia-based metal wastewater through four cycles. It could be of great potential in the practical application.

Key words: Selective biomineralization; Recovery of Au(III); AuNP; *Bacillus licheniformis* FZUL-63; Aqua regia-based metal wastewater

1. Introduction

Gold is a rare and precious metal that is widely used in various manufacturing industries such as smart phones, personal computers (PCs), and other electrical printed circuit boards (PCBs) based electronic devices due to its excellent physical & chemical properties, electrical properties, high catalytic activity and strong coordination ability[1-3]. However, the huge amounts of electronic scraps are also generated because of the technological innovations and a subsequent short lifetime of electronic devices [4-5]. PCBs are the key components and are considered as the most valuable parts among electric and electronics equipments, since they contain precious metals in higher concentrations than natural high-grade ores. For example, PCBs used in smart phones and PCs contain gold of about 280g /ton-waste, which is very high compared to 3-5 g/ton of gold in naturally occurring gold ores[5-6]. In addition, natural occurrences of these precious metals are limited and, in some mines, already depleted. Hence, to meet the increasing demand for gold, separation and recovery of gold from PCBs must be undertaken. The recovery of precious metals like gold from secondary sources is quite important from economic and environmental points of view [4, 7] .

Conventional methods such as precipitation, ion exchange, solvent extraction, and flotation for gold recovery are available, but these methods have major disadvantages like the use of toxic chemicals, high reagent requirements and generation of toxic secondary waste that required disposal[8-11]. Various biomasses have been used as biosorbent for recovery of precious metals, but they have major limitations in terms of low adsorption capacity, selectivity and reusability [12-15].

Bio-mineralization is natural phenomena and the process by which living organisms produce minerals. Bacterial mineralization is generally selective for elements[16-18]. For example, magnetotactic bacteria form magnetosomes in vivo by selectively absorbing iron in solution [19-20]. Bacterial mineralization to recovery metals has been gained a lot of attentions because of moderate reaction conditions, without toxic chemicals and good metal selectivity [21-23]. It reported that *Cupriavidus metallidurans* were responsible for the formation of secondary gold nano-mineral in the periplasmic space, which indicated that the production of secondary gold nano-mineral may be concerned with cell active reduction[24]. However, *Delftia acidovorans* induce gold ions mineralization by secreted delftibactin(a small molecule peptide) in solution[17]. Apart from the biological accumulation of AuNPs in prokaryotes, eukaryotes have also been reported to form gold nano-particles by biomineralization [23, 25-26].

This paper focuses on the development of low-cost and eco-friendly method for recovery of gold from multi-ionic aqueous systems via bacterial mineralization[12]. The *Bacillus licheniformis* FZUL-63, separated from a landscape lake in FuZhou University, was shown to selectively mineralize and precipitate gold from coexisting ions in aqueous solution. The process is as follows: Au(III) ions are reduced to monovalent by the *B. licheniformis*, and then the Au(I) was adsorbed on the bacterial surface at the beginning stage(first hour). The amino, carboxyl and phosphate groups on the surface of the bacteria are related to the adsorption of gold ions. The gold biomineralization began about 10 hours after the interaction between Au(III) ions and bacteria and has been rarely influenced by other co-existing metal ions. The formed gold nanoparticles are polyhedral structure with a particle size of ~20 nm. The bacteria could selectively mineralize and recover 478 mg/g(dry biomass) gold from aqua regia-based metal wastewater through 4 cycles. It could be of great potential in recovering Au(III) from multi-ionic aqueous systems.

2. Materials and methods

2.1. Microorganisms and growth conditions

The strain FZUL-63, isolated from a landscape lake in FuZhou University, China (GPS location: N 22°03.41 E 119°11.23), was identified through 16S rDNA sequence homology analysis based on the standard procedure[27]. The strain FZUL-63 was cultivated in LB medium with 1% Tryptone, 0.5% Yeast and 1%NaCl for 2d at 30℃. The cells were collected via centrifugation at 9,000 rpm for 10 min and the sediment was washed with 0.9% NaCl for three times, and then the cells was re-suspended in the 0.9% NaCl for the variation analysis. In order to prevent the formation of silver chloride precipitation, we use purified water instead of 0.9% NaCl to wash and re-suspend the cells that are acting with silver.

2.2. Metals ion solution and analysis

All the chemicals used in this work were of analytical grade and were obtained from Sigma-Aldrich or Aladdin Industrial Corporation. Reagent grade water with a specific resistance of 18.2 M cm was obtained from a Milli-Q water purification system. The stock solution (1,000mg/L) of Au(III), Cu(II), Pt(III), Cr(Ⅵ), Pb(II), Ag(I), Zn(Ⅱ) used in this experiments were prepared by dissolving $HAuCl_4 \cdot 3H_2O$, $CuSO_4 \cdot 5H_2O$, $HPtCl_4$, $K_2Cr_2O_7$, $PbNO_3$, $AgNO_3$ and $ZnSO_4 \cdot 7H_2O$ with purified water, respectively. The standard solution ranging from 1 to 200 mg/L of the metals ion were prepared by diluting the stock solutions. Adjustment of pH was carried out using 0.1 mol/L NaOH or 0.1 mol/L HNO3. The concentration of the metals ion was measured by ICP-OES (Optima 7000 DV, PerkinElmer, USA).

2.3. Au(III) uptake and mineralization experiments

The harvested and washed bacterial cells were transferred into a 100-mL triangular flask containing 30 mL Au(III) solution (200 mmol/L). The bacterial concentration was 0.05 mg/mL (dry weight) and pH was maintained at 7 in the initial point of the experiment. No nutrients were added to the reactors. Subsequently, the triangular flask was incubated at 30 °C with a constant shaking speed of 160 rpm. Subsamples were collected at predetermined times and centrifuged at 5400g for 10 min. The supernatant samples were filtered through 0.22-μm filters to remove Au(III) adsorbed on cell fragments. The residual Au(III) concentration in aqueous solution was determined by ICP-OES. All experiments

were performed in triplicate.

The precipitates were freeze-dried and ground to powder in a mortar. The powder samples were analyzed by XRD (MiniFlex600, Rigahu, Japan) with CuKα (λ = 0.154 nm), incident beam monochromator, and power of 40 kV × 20 mA [28]. Diffractograms were obtained over a 2θ range of 5 to 80° at a speed of 2°/min. Peak identification was achieved with a standard profile fitting routine provided by Philips Netherlands. Qualitative identification of mineral phases was made utilizing the MDI Jade 7 software[29]. The average size of the metal gold particles was calculated by Sherrer procedure.

TEM/EDS were employed to observe the distributions of Gold on the bacteria cells. After complete interaction of Au(III) (24 h of incubation), a 2-mL cell-Au(II) suspension was taken from the 200mg/L Au(III) bio-removal experiment and centrifuged at 5400g for 10 min. The resulting cell pellet was washed three times with DI water. JEOL JEM-2100 LaB6 TEM with an accelerating voltage of 200 keV fitted with STEM/EDS was employed for high resolution imaging and for compositional analysis. To identify the reduced gold mineral, selected area electron diffraction (SAED) pattern was acquired with a Gatan Orius SC200D camera[30].

Scanning electron microscopy (SEM) and elemental mapping were employed to observe the distributions of Gold on the bacteria cells. SEM observations were made to identify any morphological changes of *B. licheniformis* upon exposure of Gold(III) and to identify any mineral phase formed from Au(III) adsorption. Among the four different Au (III) concentrations used for the adsorption experiment, the 3.75 mM Au(III) sample was observed under SEM. Cells were first fixed for 20 min with 2% formaldehyde and 2.5% glutaraldehyde in a 0.05-M sodium cacodylate buffer (pH 7.2) at a 1:1 ratio (sample: fixative). After this primary fixation, a few drops of sample suspension were placed over the surface of a glass cover slip and sequentially dehydrated using varying proportions of ethanol followed by critical point drying with a Tousimis Samdri-780A critical point dryer (CPD). Critical point dried samples were coated with carbon using Denton vacuum evaporator DV-502A. A Zeiss Supra 35 VP SEM with Genesis 2000 Xray energy-dispersive spectroscopy (SEM/EDS) was employed for cell imaging and compositional analyses (Bishop et al. 2014).

2.4 X-ray photoelectron spectroscopy (XPS) analysis

The chemical states of gold in the samples were measured by XPS. The powder samples of bacteria reacting with Au(III) for different time were placed in an evacuated sample chamber. Survey spectra were collected over the range of 0～1200 eV with pass energy of 30.0 eV. High-resolution XPS spectra were acquired for C1s[31]. All binding energy values were calibrated by using the value of contaminant carbon (284.65 eV) as a reference[32]. The spectra for Au were obtained under conditions of 0.05 eV step and analyzed after corrections.

2.5 Selective Mineralization experiments

The common metal ions in electroplating wastewater include Au(III), Cu(II), Pt(III), Cr(VI), Pb(II), Ag(I), Zn(II) and so on. The above mentioned ions are prepared at every 100-mL triangular flask by diluting the stock solution, respectively. The gold stock solution is added to the various triangular flasks with the initial concentration 200mg/L except triangular flask contain the Ag(I) solution (Because Ag(I) ions cannot coexist with the $HAuCl_4 \cdot 3H_2O$ solution). After adding the Au(III)， the initial concentration of Cu(II), Pt(III), Cr(VI), Pb(II), Ag(I), Zn(II), were adjusted to 200mg/L and the bacterial dosage was 5g dry weight per liter in each 100-mL triangular flask containing 25 mL solution. Subsequently, the triangular flasks were incubated at 30 °C with a constant shaking speed of 160 rpm. Subsamples were collected at predetermined times and centrifuged at 5400g for 10 min. The residual ions concentrations in the supernatant were determined by ICP-OES and the pellet were analysis by XRD and TEM as mentioned above.

2.6 Recovery of Au(III) form aqua regia-based metal wastewater

Remove as much other metal as possible except for gold ions from the circuit printing plate with nitric acid. The insoluble substance was collected and treated with aqua regia. And then adjust pH to 6.0 with 5 mol/L KOH and centrifuge. The metal ion concentrations in the supernatant, also call it aqua regia-based metal wastewater were determined by ICP-OES as mentioned above.

The bacteria were added to the supernatant with the amount of 1 grams biomass(dry weight) per liter. Subsequently, the triangular flasks were incubated at 30 °C with a constant shaking speed of 160 rpm. Subsamples were collected at predetermined times for analysis. Reach the reaction platform stage, bacteria and gold nanoparticles were collected by centrifugation at 5400g for 10 min, and the aqua

regia-based metal wastewater was added to the collection contained bacteria and gold nanoparticles again. Cycle many times until the bacteria have less or no ability to mineralize Au(III).

3. Results and discussion

3.1 Au(III) uptake and mineralization

The strain FZUL-63 was identified as *Bacillus licheniformis* through 16S rDNA sequence homology analysis. *B. licheniformis* removed Au(III) from aqueous solution as a function of time. The Au(III) concentration in aqueous solution showed a time-course decrease, from 200 to 4.975 mg/L, and after one hour of interaction Au (III) concentration reached a steady state (Fig. 1a). In this process, the color of the culture in the triangular flask turns from bright yellow to pink (10 hours), and finally becomes wine red (Fig. 1a). The solid phase samplings collected from different interaction time were analyzed by XRD (Fig. 1b). In first hour, no peaks were detected on XRD patterns after the interaction between bacteria and Au(III), suggesting that adsorbed Au (III) did not form a crystalline phases. Although no crystalline phases were detected with XRD after one hour interaction, with prolonged time, some weak peaks were observed on XRD pattern after 10 hours of interaction, implying that a small amount of gold mineral formed (Fig. 1b). The newly observed diffraction peaks matched well with gold nanoparticles [Au(PNs)]. These results suggest that with time the bacterium may have mediated Au (III) transformation from amorphous compound to a stable AuNPs. Under TEM, there were many AuNPs with a ~20 nm grain size on and around bacterial cell surface (Fig. 1a).

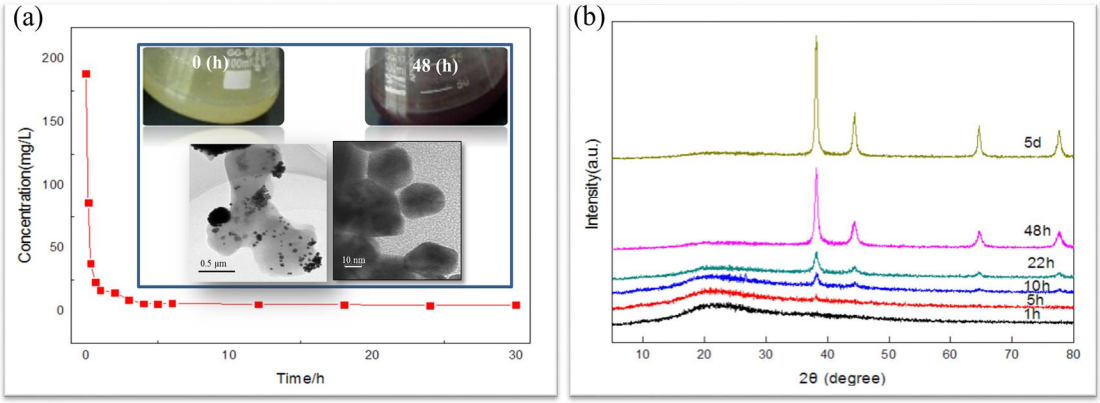


Fig 1.The interaction between bacteria and Au(III) with time. (a). The Au(III) concentration in aqueous solution showed a time-course decrease. (b). XRD pattern of the Au(PNs) enhanced over time.

3.2 The chemical groups involved in gold binding

The gold ions in the aqueous solution is quickly removed (Fig. 1a), but the XRD results indicate that no minerals have been formed at the initial stage (Fig. 1b), so the gold ions may be adsorbed on the surface of the bacteria first. To elucidate the chemical groups involved in gold binding, FTIR spectra were recorded for control and gold loaded cells from 4000 cm^{-1} to 400 cm^{-1} (Fig. 2).

The characteristic peaks can be assigned to the involvement of the main functional groups present in the bacterial biomass by analyzing the highly complex IR spectra (Fig. 2). The N–H stretching peak lies in the spectrum region occupied by a broad and strong band in 3200–3600 cm^{-1} region was due to the presence of γ O–H of the hydroxyl groups, which undergo change in peak position in gold loaded spectrum suggesting the involvement of amino and hydroxyl groups in gold binding to bacterial surface[33-34]. In the spectra for gold loaded cell the peak around 2366 cm^{-1} could be assigned to the P–O stretching vibrations implying possible role of phosphate metabolism facilitating cellular adsorption of gold ions. The spectra for both control and gold loaded samples revealed protein related bands. The appearance of γ C=O of amide I and δ NH/ γ C=O combination of the amide II bonds at were present at 1652 cm^{-1} and 1543 cm^{-1}, respectively were predominant in the control spectrum. Following Au binding, the amide I absorption peak (1652 cm^{-1}) was split in to two minor peaks at 1664 cm^{-1} and 1639 cm^{-1} and a marked shift of 1543 cm^{-1} peak to 1541 cm^{-1} suggest a strong interaction of Au with carboxyl groups. In the control spectrum, sharp peaks in between 1400 cm^{-1} and 1500 cm^{-1} were due to the presence of the carboxyl groups [31]. Particularly, the strong peak at 1451 cm^{-1} which was characteristic of the scissoring motion of CH2 groups [32] undergo a shift to lower energy level (1449 cm^{-1}) after gold binding. Following gold uptake a clear shift of the peak at 1399 cm^{-1} to 1388 cm^{-1} due to symmetric stretching of COO– vibration strongly indicated role of carboxyl groups in gold binding[34]. In the control spectrum, the strong peaks at 1238 cm^{-1} and 1062 cm^{-1} were observed due to vibrations of carboxyl and phosphate groups[35]. Following gold exposure, a clear shift of these peaks to 1234 cm^{-1} and 1057 cm^{-1} suggests interaction of bound metals with carboxyl and phosphates groups. A gradual shift of the peak in control spectra at 1239 cm^{-1} because of asymmetric stretching modes of protonated polyphosphates and PO uncomplexed in phosphate diesters to 1238 cm^{-1} in gold loaded samples indicated the weakening of the P=O character as a result of metal binding to the phosphates[35]. Changes in peak position and intensity at 800 cm^{-1} to 400 cm^{-1} region could be assigned to the formation of intense (M–O) and (O–M–O) bonds (M =metal ion)[33]. The overall IR spectroscopic analysis suggests that phosphate, carboxyl and amide groups on bacterial cell are the

dominant functional groups involved in bacteria-gold interaction.

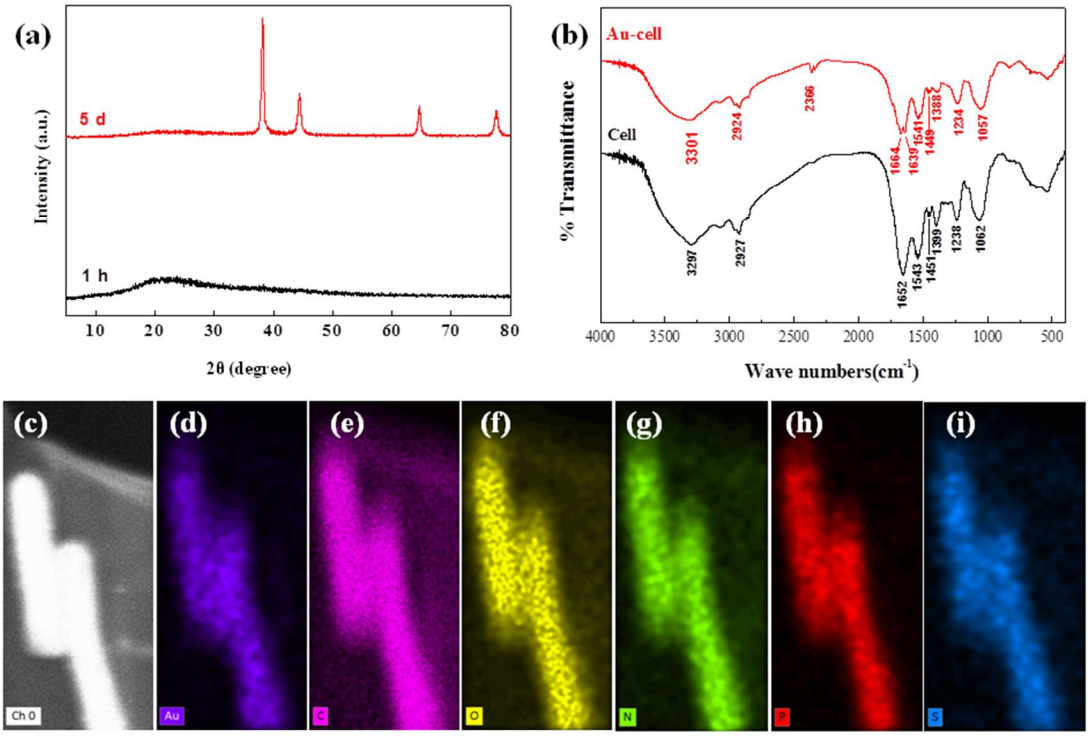
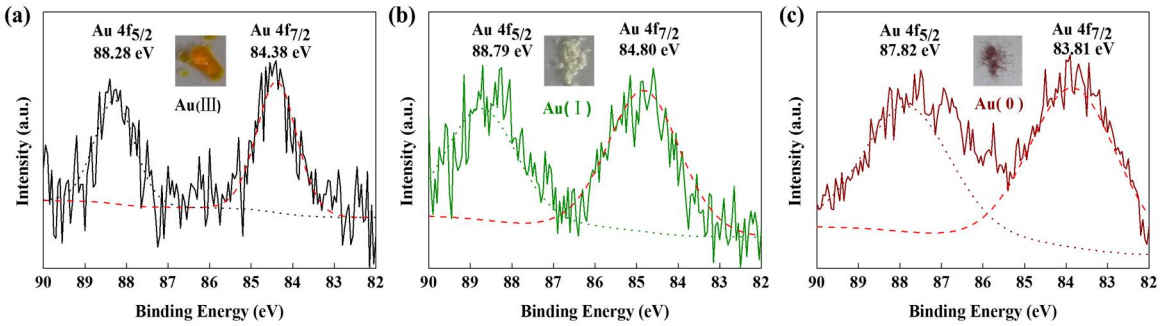


Fig. 2. Fourier transformed Infra Red spectra of *B. licheniformis* FZUL-63 biomass: before and after gold uptake (initial concentration 200 mg Au (III) L^{-1} at pH 6.0, time 1 h).

3.3 Changes in the chemical valence state of Gold

The interaction of Au(III) with bacteria at different time have been investigated by XPS. The Au 4f spectra of all the investigated samples are shown in Fig. 3. The evidence is confirmed by the curve-fitting of the Au 4f core-level spectra by two spin-orbit splitted Au 4f7/2 and Au 4f5/2 components (DE：~ 4.0 eV), showing increasing BE values that correspond to a different chemical state of gold particles in the samples. In the initial addition of chloric acid sample the Au 4f7/2 photoelectronic peak is located at BE = 84.38 eV and this value is typical of Au(III) species Fig. 3a. After the interaction of the trivalent gold ions with bacteria, in the one hour sample shown in Fig. 3b and the Au 4f 7/2 spectrum consists of only one components located at BE = 84.80 eV, which can be assigned to Au(I) species. In the 48 hours sample the Au 4f7/2 peak detected at BE = 83.81 eV shown in Fig. 3c can be attributed to the presence of Au(0) species on the bacteria surface. From the changes in the valence state after the action of trivalent gold ions and bacteria, it could be deduced that trivalent gold ions are not reduced to zero-valent gold(AuNP) in one step and the intermediates of monovalent gold compounds were

225 produced in the reduced process.



226 Fig. 3 XPS spectra of Au(4f) region of interaction with bacteria for 0 h(a), 1h (b) and 48h(c).

227

228 **3.3 Coexisting ions affect the mineralization and recovery of gold**

229 The aqua regia-based metal wastewater usually contains the following metal ions such as Au(III),
230 Pt(III), Ag(I), Cr(Ⅵ), Pb(Ⅱ), Zn(II), Cu(II). The chloroauric acid and the silver ion cannot coexist in
231 the solution. Therefore, we only did experiments using gold and silver acting alone with bacteria, their
232 recovery rates were 97.66 and 49.23, respectively (Fig 4 a). The 200 mg/L Cr(Ⅵ) ions could delay the
233 recovery balance time of the gold from 1 hours to 18 hours, the recovery rate of gold was also reduced
234 from 97.66% to 91.93% (Fig 4 b). Zn(Ⅱ) and Cu(Ⅱ) almost rarely influenced the Au(III) recovery
235 (Fig 4 c, d). However, when the 200 mg/L Pt(III) or Pb(Ⅱ) coexisting with the Au(III), the recovery
236 rate of gold was reduced from 97.66% to 85.06% and 93.31%, respectively (Fig e, f).

237 On the contrary, when these ions coexist with the gold ions, the gold ions could competitively
238 reduce the adsorption of bacteria to these co-existing ions, especially Pt and Pb ions have obvious
239 reduction. Compared with the removal rate of Pt(III) and Pb(II) via bacteria alone, the removal rate of
240 Pt(III) and Pb(II) decreased from 45.82% to less 0.5% and 56% to17.33% in the presence of gold ions,
241 respectively (Fig 4. c, d). This facilitates the selective recovery of gold from multi-ionic aqueous
242 systems.

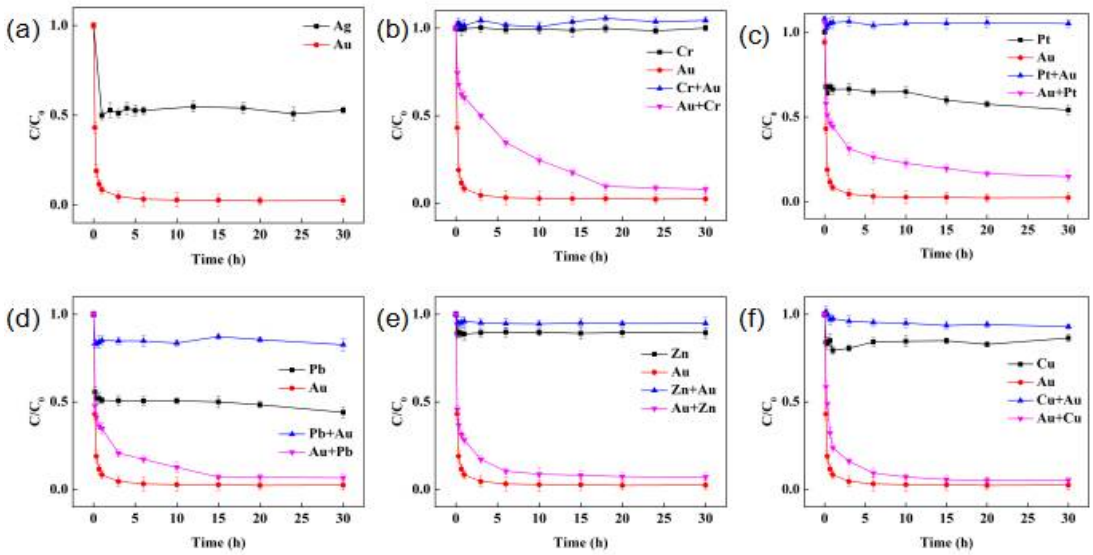


Fig 4. Au(III) recovery influenced by coexist metal ions. The initial concentration of each ion is 200mg/L, and the bacterial dosage is 1g dry weight per liter.

3.4 Recovery of Au(III) form metal wastewater

The CPBs is treated with strong nitric acid. The insoluble substance was collected by centrifugation and treated with aqua regia. And then adjust pH to 6.0 and centrifuge. The concentration of ions in the supernatant was measured by ICP, and the concentration of each ion before and after bacterial treatment was shown in table 1.

Table 1 Recovery of Au(III) from aqua regia-based metal wastewater at first cycle.

Metal concn. mg/L	Au^{3+}	Cu^{2+}	Zn^{2+}	Pb^{2+}	Pt^{4+}	Cr^{6+}	Fe^{3+}	Ni^{2+}	Ag^{+}
Without Cell	181.2±3.2	348.5±3.7	120.0±2.3	39.2±2.6	16.1±1.1	27.3±2.4	85.4 ±3.5	48.7±2.9	ND
With Cell	17.4±1.2	330.7±2.5	115.2±2.2	34.3±1.7	15.9±1.3	26.1±1.9	80.6±3.1	47.3±2.7	ND
Removal efficiency (%)	90.4%	5.1%	4.0%	12.5%	1.3%	4.4%	5.6%	2.9%	--

Note: ND stands for not detected. All experiments were performed in triplicate.

As the Fig 4 show, through the first treatment, the recovery rate of gold was 90.4%. After the second treatment, the gold recovery dropped to 82.3%. The gold recovery rates were 61.8% and 29.6% at third and fourth treatment, respectively. With four cycles, we can recover 478 mg of gold per gram of bacteria from the aqua regia-based metal wastewater.

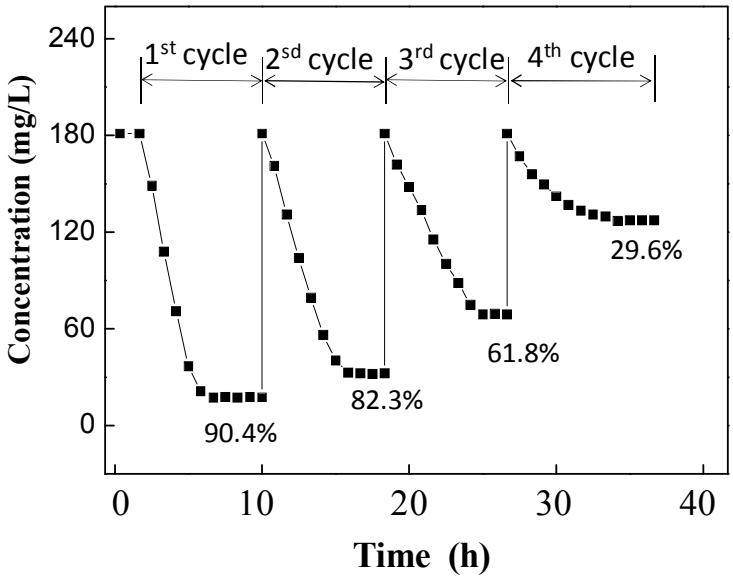


Fig 4. The *Bacillus licheniformis* FZUL-63 selectively mineralizes and recovers gold from aqua regia-based metal wastewater through four cycles.

4. Conclusion

An indigenous bacterium *Bacillus licheniformis* FZUL-63 was shown to selectively mineralize and precipitate gold from coexisting ions in aqueous solution. The process is as follows: Au(III) ions are reduced to monovalent by the *B. licheniformis*, and then the Au(I) was adsorbed on the bacterial surface at the beginning stage. The amino, carboxyl and phosphate groups on the surface of the bacteria are related to the adsorption of gold ions. The gold biomineralization has been rarely influenced by other co-existing metal ions. The bacteria could recover 478 mg/g(dry biomass) gold from aqua regia-based metal wastewater through 4 times.

Acknowledgments

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