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Bioleaching of sludge from acid-leached waste traction batteries used in electric vehicles for the extraction of Ni and Co using optimized microbial consortia

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Bioleaching of heavy metals from sludge from acid-leached waste lithium batteries is limited by the restricted maximum acid-producing ability of a single bacterium. Compatibility of acidophilic autotrophic bacteria in bioleaching is desirable. Herein, five microorganisms, including *Acidithiobacillus ferrooxidans* (Af), *Leptospirillum ferriphilum* (Lf), *Thiobacillus thiooxidans* (Tt), *Acidithiobacillus caldus* (Ac), *Sulfobacillus thermotolerans* (St) and their mixed microbial consortia (MC), were applied to leach Ni and Co from sludge from acid-leached waste lithium batteries. Bioleaching kinetics were investigated as functions of single bacterium species, MC species, Ac and St ratios in MC, and solid/liquid ratio. Compared with the single bacterium, the bioleaching ratios of MC with 20% of Ac and 20% of St (45 °C and a solid/liquid ratio of 10%) were 1.17 times higher for Ni (bioleaching ratio of 88.4% and equilibrium time of 8 days), and MC with 25% of St (40 °C and solid/liquid ratio of 10%) presented 1.18 times higher bioleaching efficiency for Co (bioleaching ratio of 75.4% and equilibrium time of 6 days). The quantitative ratios of Ni and Co species, including acid-soluble, reducible, oxidizable, and residual states in the remaining bioleached sludge, were determined. MC presented optimal bioleaching efficiency with 90.3% for the leached fraction and 5.9% for the residual fraction for Ni, and 82.9% for the leached fraction and 11.8% for the residual fraction for Co. This study provides a valuable reference for efficiently bioleaching waste lithium batteries.

1. Introduction

The number of electric vehicles is increasing rapidly, resulting in a concomitant increase in discarded power batteries. In 2021, the theoretical amount of battery recycling for electric vehicles reached 294 000 tons, accounting for half of the total amount of discarded batteries in China.[1] Lithium battery waste is expected to reach 11 million tons by 2030.[2] Meanwhile, it is estimated that waste lithium-ion battery recycling in China will reach 2.312 million tons in 2026. Accordingly, the problem of recycling waste lithium batteries is a significant concern. Heavy metals such as lithium, cobalt, manganese and nickel in cathode materials in waste lithium battery residuals have significant negative influences on soil, water and the atmospheric environment.[3] For instance, the released lead in waste lithium batteries penetrates into the soil, resulting in the permanent loss of use of the soil and affecting the plant growth and the health of the ecosystem.[4,5] A single button cell could pollute 600 000 liters of water, which is equivalent to the one person's lifetime drinking water.[6] The combustion of waste lithium batteries produces harmful gases, including carbon dioxide, carbon monoxide and hydrogen sulfide, which affect both the air quality and human health. Therefore, it is urgent to manage the residual heavy metals in the cathode materials of waste lithium batteries.

Treatment methods for waste lithium batteries mainly include mechanical treatment, pyrometallurgy, hydrometallurgy and bioleaching.[7–11] Recently, bioleaching has been widely used in the leaching of metal-containing electronic waste because of its simple operation, low cost, minimum energy input, reduced demand for skilled labor and environmental friendliness, which can widely reduce pollution and greenhouse gas emissions.[12–15] Metal elements can be effectively separated from non-metal elements in solid waste and then released into the solution system.[16–19] Previous studies presented that the selection of suitable microorganisms plays a decisive role in the final metal bioleaching efficiency. Widely used bioleaching microorganisms include *Thiobacillus ferrooxidans*, *Thiobacillus thiooxidans* and *Leptospira ferrooxidans*.[20–22] They can oxidize low-valent iron and sulfur elements into trivalent iron and sulfuric acid, promote the formation of minerals in the presence of a suitable nitrogen source, reduce the pH value of the

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system, and generate high redox potential.[23,24] So far, the existing research on biological leaching of heavy metals in solid wastes by acidophilic autotrophic bacteria is mainly based on a single dominant bacterium.[25] The limited maximum acid-producing capacity and single acid-producing type of individual bacteria make it impossible to ensure the timely and effective removal of the diverse kinds and forms of heavy metals from solid wastes during biological leaching using just one bacterium. Therefore, using combinations of acidophilic autotrophic bacteria with a wide acid-producing spectrum and high acid-producing efficiency is a possible approach for improving the technical and economic feasibility of bioleaching. Zhou *et al.* presented a two-stage leaching strategy (*i.e.*, bioleaching combined with brine leaching) as a feasible and promising treatment for multi-metal removal from plant ash.[26] Moreover, since acid leaching is a common hydrometallurgical technique for metal recovery from solid waste, the combination of acid leaching and bioleaching could improve the leaching efficiency to the maximum from a practical view. For the acid-leached sludge produced by factories, a more efficient metal-extraction technique from acid-leaching metal-containing sludge using mixed bacterial bioleaching has not yet been described.

In this study, five bacteria, including *Acidithiobacillus ferrooxidans* (Af), *Leptospirillum ferriphilum* (Lf), *Thiobacillus thiooxidans* (Tt), *Acidithiobacillus caldus* (Ac) and *Sulfobacillus thermotolerans* (St) and their mixed microbial consortium (MC) were applied to bioleach sludge of acid-leached waste lithium batteries for efficient release of Ni and Co. The bioleaching kinetics for Ni and Co were investigated as functions of single bacterium species, MC species, Ac and St ratio in MC, and solid/liquid ratio. Moreover, the Ni and Co species, including acid-soluble, reducible, oxidizable, and residual states in the sludge remaining from bioleached traction batteries and the leached fraction, were determined.

2. Materials and methods

2.1. Solid waste materials

The tailings were produced by a domestic leading recycling enterprise in wet production lines. Waste power battery cathode materials and other recycled wastes were crushed and sorted. Then they entered independent wet recovery production lines to leach Ni and Co, producing Ni slag and Co slag, respectively. The concentrations of Ni in the Ni slag and Co in the Co slag were measured using a handheld X-ray fluorescence (XRF) spectrometer, as shown in Table S1. The measured concentration of Ni in the Ni slag was 0.8214%, and that of Co in the Co slag was 0.4998%.

2.2. Microorganisms and cultivation

Five typical bioleaching strains, including *Acidithiobacillus ferrooxidans* (Af, ATCC 23270), *Leptospirillum ferriphilum* (Lf, AS1 6358), *Thiobacillus thiooxidans* (Tt, BJMCC 01101), *Acidithiobacillus caldus* (Ac, CGMCC 1.15711) and *Sulfobacillus thermotolerans* (St, DSM 17362) and their mixed microbial consortia (MC) were used. These five bacteria were cultured in improved 9K medium (pH = 1.9). The composition of the modified 9K medium was as follows: $(NH_4)_2SO_4$ 3.0 g L^{-1}, KCl 0.1 g L^{-1}, K_2HPO_4 0.5 g L^{-1}, $MgSO_4 \cdot 7H_2O$ 0.5 g L^{-1}, and $Ca(NO_3)_2$ 0.01 g L^{-1}.[27] $FeSO_4 \cdot 7H_2O$ (44.7 g L^{-1}) was supplemented to Af, Lf and St as an energy source. Sulfur powder (5 g L^{-1}) was supplemented to Tt as an energy source. Sulfur powder (20 g L^{-1}) was supplemented to Ac as an energy source. MC was also cultured in an improved 9K medium (pH = 1.9) with the addition of sulfur powder and ferrous sulfate heptahydrate as optimized energy sources.

2.3. Bioleaching experiments

2.3.1. Bioleaching using a single bacterium. The bioleaching of Ni and Co from acid-leached slags was carried out using single bacterial species, including Af, Lf, Tt, Ac, St and a blank group (CK). The bioleaching experiment was carried out in a 3 L glass beaker, and a 2 L leaching system (9K medium, pH 1.9) was set up, which contained 200 mL of bacterial liquid. After the pre-culture, the sample was added for the biological leaching experiment. The beaker was stirred on a constant-temperature magnetic stirrer to fully suspend the waste residue, the suspension position was marked, and the evaporated water was replenished every day. Sampling method for the leaching test: during the experiment, 5 mL samples of the suspension were taken, filtered and mixed with nitric acid for preservation. Initially, samples were taken at the 2nd hour, 6th hour and 12th hour. After that, sampling should be done every other day. From the 4th day, sampling should be done every two days until the 12th day. The bioleaching temperature for the Af, Lf, Tt and CK groups was set at 30 °C, and for Ac and St it was set to 40 °C. The mass content of Ni and Co in the liquid for all samples was tested using inductively coupled plasma-mass spectrometry (NexION 2000, PerkinElmer). The solid/liquid ratio for the bioleaching experiment was 10%.

2.3.2. Bioleaching experiments using MC type. The bioleaching of Ni and Co from acid-leached slags was carried out using MC. Six different microbial consortiums (as listed in Table 1) were obtained by mixing equal volumes of the single bacteria. The biological leaching experiment was carried out at different temperatures (30 °C, 40 °C and 45 °C) with a solid/liquid ratio of 10%.

2.3.3. Bioleaching experiments with various Ac and St ratios in MC. The microbial leaching of Ni and Co from slags was carried out by increasing the proportion of Ac and St as listed in Table 2. For the bioleaching of Ni from Ni slag, MC45-

Table 1 Naming of bacterial MC specimens with various single bacterium types

MC specimen	Temp.	Abbreviation
Af, Lf and Tt with the same volumes	30 °C	MC30-0AS
Ac and St with the same volumes	40 °C	AS
Af, Lf, Tt and Ac with the same volumes	40 °C	MC40-25Ac
Af, Lf, Tt and St with the same volumes	40 °C	MC40-25St
Af, Lf, Tt, Ac and St with the same volumes	40 °C	MC40-40AS
Af, Lf, Tt, Ac and St with the same volumes	45 °C	MC45-40AS

Table 2 Naming of bacterial MC specimens with various Ac and St ratios

MC specimen	Temp.	Abbreviation
20% Af, 20% Lf, 20% Tt, 20% Ac, 20% St	45 °C	MC45-40AS
16.67% Af, 16.67% Lf, 16.67% Tt, 25% Ac, 25% St	45 °C	MC45-50AS
8.33% Af, 8.33% Lf, 8.33% Tt, 37.5% Ac, 37.5% St	45 °C	MC45-75AS
25% Af, 25% Lf, 25% Tt, 25% Ac	40 °C	MC40-25Ac
16.67% Af, 16.67% Lf, 16.67% Tt, 50% Ac	40 °C	MC40-50Ac
8.33% Af, 8.33% Lf, 8.33% Tt, 75% Ac	40 °C	MC40-75Ac
25% Af, 25% Lf, 25% Tt, 25% St	40 °C	MC40-25St
16.67% Af, 16.67% Lf, 16.67% Tt, 50% St	40 °C	MC40-50St
8.33% Af, 8.33% Lf, 8.33% Tt, 75% St	40 °C	MC40-75St

40AS was used as the basis leaching bacteria with increasing ratios of Ac and St. For the bioleaching of Co from Co slag, MC40-25Ac and MC40-25St were used as the basis leaching bacteria with improving ratios of Ac and St. The solid/liquid ratio was set to 10%.

2.3.4. Bioleaching experiments with various solid/liquid ratios. The most suitable bioleaching MC for Ni and Co from slags were selected from the above experiments. MC45-40AS was the optimal bioleaching MC for Ni and MC40-25St was the optimal bioleaching MC for Co. Experiments were carried out to evaluate the bioleaching with different solid/liquid ratios. The solid/liquid ratio was set to 10%, 20%, 30%, 40% and 50%. Samples of 200, 400, 600, 800 and 1000 g were added to the 2 L leaching system for biological leaching experiments.

2.4. Determination of Ni and Co species

The improved four-step extraction method recommended by the European Community Bureau of Reference (BCR) was applied to determine the Ni and Co species in bioleached slags.[28,29] It mainly includes the following four parts. (1) Acid-soluble fraction (F1). First, 0.5 g of a sample was added into a 50.0 mL centrifuge tube, and 20.0 mL 0.11 mol per L CH_3COOH was added for leaching and oscillated at 180 rpm at room temperature of 25 °C for 16 hours. The mixture was centrifuged at 3000 rpm for 20 min, and the supernatant obtained by filtration was the acid-soluble fraction, which was placed in a refrigerator at 4 °C for testing. (2) Reducible fraction (F2). To the solid residue obtained after the F1 extraction was added 20.0 mL of 0.5 mol per L $NH_2OH\cdot HCl$, which was shaken at room temperature of 25 °C at 180 rpm for 16 hours and centrifuged at 3000 rpm for 20 min. The supernatant obtained by filtration was F2, and it was placed in a refrigerator at 4 °C for testing. (3) Oxidizable fraction (F3). 5.0 mL of 30% H_2O_2 (pH = 2.0) was added to the remaining solid of F2 with a cover, and kept at room temperature for 1 hour. Then the mixture was put in a water bath at 85 °C for 1 hour. The cover was removed and continue to digest until 3.0 mL of the liquid remained. Then 5.0 mL of H_2O_2 was added to cover it, and heating continued for 1 hour. The cover was opened and the mixture was evaporated until the liquid was about 1.0 mL. After cooling to room temperature, 25.0 mL of 1.0 mol per L CH_3COONH_4 was added, then the mixture was shaken at 25 °C for 16 hours at 180 rpm, and centrifuged at 3000 rpm for 20 min. The supernatant was filtered to give F3, and put in a refrigerator at 4 °C for testing. (4) Residue fraction (F4). The dried F3 residue was taken in a sand bath with mixed acid by digestion method for determining the total amount of heavy metals.

3. Results and discussion

3.1. Bioleaching of Ni and Co from sludge of acid-leached waste lithium batteries

The microbial community dynamics of the suspended microorganisms during the cultivation process are crucial for the bioleaching of metal ions from sludge. According to previous studies reported by Zhou *et al.*, it takes 6 days to reach a steady concentration for three kinds of bacteria including *Leptospirillum ferriphilum* CS13, *Sulfobacillus acidophilus* CS5 and *Acidithiobacillus caldus* S2.[30] Zhou's previous experimental research also found that it took more than 60 h for moderately thermophilic microorganisms to complete a batch of cultivation, and after the 30 days of batch cultivation, the amount of biomass obtained was only half of the continuous cultivation in this experiment.[31,32] Thus, the single bacterium and various types of MC were cultivated for 7 days and used directly in this work.

Moreover, pH was crucial in the bioleaching process due to the promotion of bioleaching by protons. Zhou *et al.* also reported the effect of pH on heavy metal leaching rates in the bioleaching process.[30] According to their studies, pH < 2.0 would be appropriate for the efficient bioleaching of heavy metals from sludge. Considering the amount of acid used in practical application and the bioleaching efficiency, a pH of 1.9 for the bioleaching systems was applied in this study.

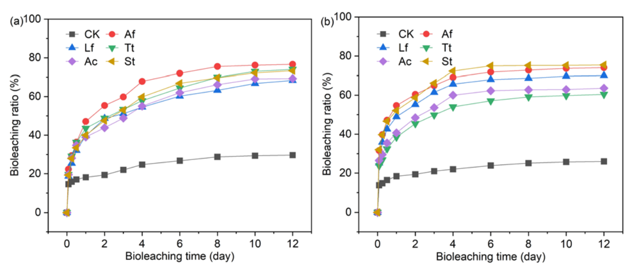

Fig. 1 Bioleaching kinetics as a function of single bacteria species, including CK, Af, Lf, Tt, Ac and St from Ni slag (a) and Co slag (b).

3.1.1. Bioleaching kinetics as a function of single bacterium. The bioleaching efficiency of Ni from sludge from acid-leached waste lithium batteries is shown in Fig. 1a. Without the addition of any bacterium, the bioleaching ratio reached equilibrium with a low value of 28.7% and a slow time of 8 days. The bioleaching ratio for Lf reached equilibrium with a low value of 66.7% and a slow time of 10 days. The bioleaching ratios for the other four bacteria reached equilibrium at 8 days. Specifically, their equilibrium ratios were 75.5% for Af, 66.1% for Ac, 69.7% for St and 69.9% for Tt. It is indicated that Af presented the highest bioleaching ratio and fastest equilibrium compared with the other single bacteria.

The bioleaching efficiency of Co from the sludge from acid-leached waste lithium batteries is shown in Fig. 1b; the bioleaching ratios with the addition of Ac, St, Af and Lf bacteria could be reached at 4 days with ratio values of 60.1% for Ac, 72.3% for St, 69.1% for Af and 65.6% for Lf. With the addition of Tt, the bioleaching equilibrium could be achieved at 8 days with a ratio of 54.0%, which was significantly slower than other bacteria. It is indicated that St presented the highest bioleaching ratio and fastest equilibrium compared with other single bacteria. Thus, MC with various ratios of the bacteria would be applied as the basis bacteria for Ni and Co to evaluate the effects of the different bacteria species in MC in the following experiment.

3.1.2. Bioleaching kinetics as a function of MC type. The bioleaching kinetics of Ni and Co from the sludge from acid-leached waste lithium batteries by various MC types, including MC30-0AS, AS, MC40-25AS, MC40-25St, MC40-40AS, and MC45-40AS, are shown in Fig. 2. As presented in Fig. 2a, the Ni bioleaching ratios for six types of MC increased continuously and reached equilibrium at 8 days. The equilibrium values were 80.7% for MC30-0AS, 78.4% for AS, 83.6% for MC40-25AS, 82.4% for MC40-25St, 84.5% for MC40-40AS and 88.4% for MC45-40AS. MC45-40AS exhibited the highest bioleaching ratio for Ni compared with the other five types of MC and single bacterium species. The bioleaching of Co is presented in Fig. 2b; the bioleaching ratios for the six types of MC rose continuously and reached equilibrium at 6 days. The equilibrium ratio values were 75.4% for MC30-0AS, 68.9% for AS, 76.6% for MC40-25AS, 79.6% for MC40-25St, 70.8% for MC40-40AS and 74.4% for MC45-40AS. Thus, MC40-25St displayed the highest bioleaching ratio for Co compared with the other five types of MC and single bacterium species. Moreover, the compound consortium was more appropriate for leaching Ni and Co from the sludge from acid-leached waste lithium batteries. Hence, MC with mixed single bacterium of AC and St would be applied as the basis bacteria for Ni and Co to evaluate the ratios of AC and St in the following experiment.

3.1.3. Bioleaching kinetics as a function of Ac and St ratios in MC. As mentioned above, the optimal MC type for Ni bioleaching from the sludge from acid-leached traction batteries was MC45-40AS, and the optimal MC types for Co bioleaching from sludge from acid-leached traction batteries were MC40-25Ac and MC40-25St. It would be desirable to turn the detailed proportions of mesophilic bacteria (*i.e.*, Ac and St) in MC. As shown in Fig. 3a, MC containing three volume concentrations of Ac and St, including MC45-40AS, MC45-50AS and MC45-75AS, were applied to bioleach Ni from sludge. The equilibrium ratio values were 88.4% for MC45-40AS, 82.9% for MC45-50AS and 80.7% for MC45-75AS, indicating that MC45-40AS with 20% of Ac and 20% of St was more appropriate to bioleach Ni. As shown in Fig. 3b, MC40 containing three volume concentrations of Ac (25%, 50% and 75%) and three volume concentrations of St (25%, 50% and 75%) were applied to bioleach Co from sludge. The equilibrium ratio values were 76.6% for MC40-25Ac, 75.3% for MC40-50Ac, 71.6% for MC40-75Ac, 79.6% for MC40-25St, 78.7% for MC40-50St, and 72.4% for MC40-75St. Increasing the ratio of Ac or St in MC from 25% to 75% was not favorable for the bioleaching. Moreover, the equilibrium times for the bioleaching of Ni and Co from the sludge from acid-leached waste lithium batteries were not impacted by changing the concentrations of Ac and St in MC. Therefore, MC45-40AS and MC40-25St would be applied as the basis bacteria for Ni and Co to evaluate the solid/liquid ratio MC in the following experiment.

3.1.4. Bioleaching kinetics as a function of solid/liquid ratio. There is an equilibrium between the dissolved heavy metal ions in the solution and the heavy metals in the solid. Before arriving at the threshold value, a higher solid/liquid ratio would induce a higher concentration of heavy metals to be extracted into the liquid, which is conducive to the equilibrium moving in the direction of leaching, thus making the lower residual heavy metals in the solid as well as improving the leaching rate. Herein, five solid/liquid ratios, including 10%,

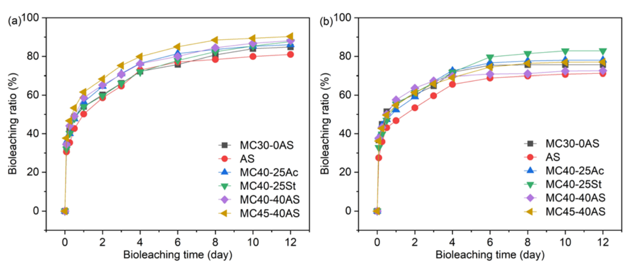

Fig. 2 Bioleaching kinetics of Ni (a) and Co (b) from the sludge from acid-leached waste lithium batteries as a function of MC type, including MC30-0AS, AS, MC40-25AS, MC40-25St, MC40-40AS, and MC45-40AS.

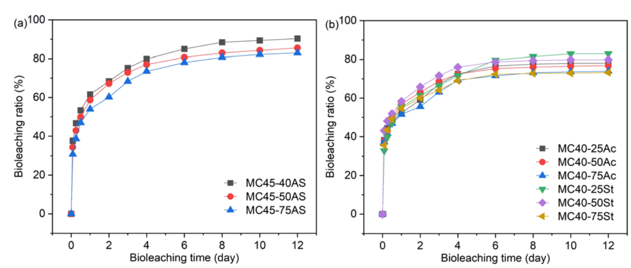

Fig. 3 Bioleaching kinetics of Ni (a) and Co (b) from the sludge from acid-leached waste lithium batteries as a function of Ac and St ratios in MC.

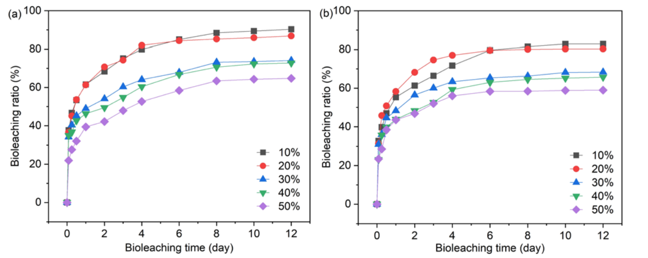

Fig. 4 Bioleaching kinetics as a function of solid/liquid ratio for Ni by MC45-40AS (a) and Co by MC40-25St (b).

20%, 30%, 40% and 50% of samples, were applied to leach Ni and Co, respectively. As shown in Fig. 4a, the equilibrium ratio values of Ni from the sludge from acid-leached waste lithium batteries were 88.4% for 10% of sample, 85.2% for 20%, 73.2% for 30%, 70.6% for 40% and 63.5% for 50%. The equilibrium ratio values of Co from the sludge were 82.9% for 10% of sample, 80.3% for 20%, 68.3% for 30%, 65.7% for 40%, and 58.9% for 50%. Thus, a solid/liquid ratio of 10% was optimal for the bioleaching of Ni by MC45-40AS and the bioleaching of Co by MC40-25St. Moreover, the equilibrium times for bioleaching Ni and Co from the sludge from acid-leached waste lithium batteries were not influenced by varying the solid/liquid ratio of the MC.

3.2. Determination of Ni and Co speciation in bioleached sludge

3.2.1. Ni speciation determination. Ni species in bioleached sludge were determined using BCR analysis as shown in Fig. 5. Typically, there were five species for Ni, including leached, residual, oxidizable, reducible and acid-soluble species. The bioleaching BCR analysis using a single type of bacterium is illustrated in Fig. 5a. Without the addition of bacteria, the leached and residual fractions for Ni in bioleached sludge were 30.9% and 58.6%, respectively. For the bioleaching of Ni using a single bacterium, the leached percentages were 76.6% for Af, 68.3% for Lf, 74.0% for Tt, 69.2% for Ac and 73.3% for St, which were obviously higher than that of the CK group. The residual percentages were 18.4% for Af, 26.8% for Lf, 23.5% for Tt, 22.7% for Ac and 19.6% for St, which were significantly lower than that of the CK group. The fractions of the other three species ranged from 0.6% to 4.5%, which demonstrated that the major fractions of Ni in bioleached sludge have been leached and transformed by the single bacterium species. The bioleaching BCR analysis using various types of MC is illustrated in Fig. 5b. The bioleaching efficiency was enhanced distinctly. Specifically, the leached fractions were 84.8% for MC30-0AS, 80.9% for AS, 86.0% for MC40-25AS, 87.6% for MC40-25St, 88.1% for MC40-40AS, and 90.3% for MC45-40AS. The residual fractions were 13.3% for MC30-0AS, 15.8% for AS, 11.8% for MC40-25AS, 10.6% for MC40-25St, 9.5% for MC40-40AS, and 5.9% for MC45-40AS. The fractions of the other three species ranged from 0.4% to 1.8%. Thus, it is verified that the residual fractions of Ni in bioleached sludge when adding MC were much less than those when adding a single bacterium. The lowest fraction value presented by MC45-40AS was also consistent with the highest bioleaching efficiency of MC45-40AS discussed in the above sections. The bioleaching BCR analysis of Ni in the bioleached sludge using various Ac and St ratios in MC45-40AS is shown in Fig. 5c. The bioleaching efficiency was enhanced distinctly. Specifically, the leached fractions were 85.5% for MC45-50AS and 82.9% for MC45-75AS. The residual fractions were 9.7% for MC45-50AS and 11.9% for MC45-75AS. The fractions of the other three species ranged from 1.8% to 2.4%. Moreover, the BCR analysis of Ni in sludge bioleached using various solid/liquid ratios is shown in Fig. 5d. As the solid/liquid ratio of MC45-40AS increased from 10% to 50%, the leached fraction of Ni decreased continuously from 90.3% to 65.8%, while the residual fraction increased monotonously from 5.9% to 19.3%. Taken together, the above results show

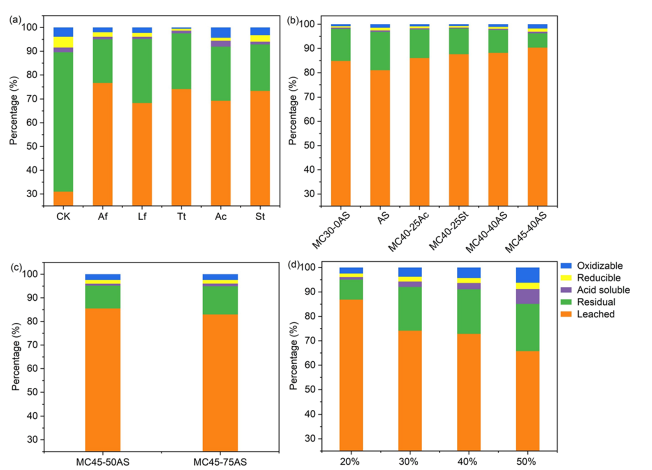

Fig. 5 Determination of Ni speciation in bioleached sludge using various types of bacteria: single bacterium (a), MC with different types (b), MC with different contents of AS (c) and different solid/liquid ratios (d).

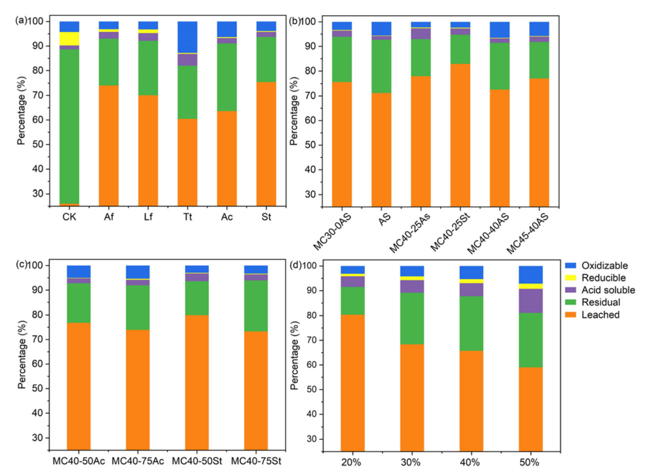

Fig. 6 Determination of Co speciation in bioleached sludge using various types of bacteria: single bacterium (a), MC with different types (b), MC with different contents of AS (c) and different solid/liquid ratios (d).

that MC45-40AS is the optimal adapted microbial consortium for the efficient bioleaching of Ni from bioleached sludge.

3.2.2. Co speciation determination. The bioleaching BCR analysis of Co species in sludge bioleached by a single type of bacterium is illustrated in Fig. 6a. Without the addition of any bacterium, the leached and residual fractions for Co in bioleached sludge were 26.0% and 62.6%, respectively. When the single bacterium was added separately, the leached percentages were 74.0% for Af, 70.0% for Lf, 60.4% for Tt, 63.6% for Ac and 75.4% for St, which were obviously higher than that of the CK group. The residual percentages were 18.9% for Af, 22.1% for Lf, 21.5% for Tt, 27.4% for Ac and 18.3% for St, which were significantly lower than that of the CK group. The fractions of the other three species ranged from 0.3% to 12.8%. Thus, the major fractions of Co species in bioleached sludge were leached by adding single bacterium species. The bioleaching BCR analysis of Co with six different types of MC is shown in Fig. 6b. The leached fractions were 75.6% for MC30-0AS, 71.2% for AS, 78.0% for MC40-25AS, 82.9% for MC40-25St, 72.6% for MC40-40AS, and 77.1% for MC45-40AS. The residual fractions were 18.3% for MC30-0AS, 21.5% for AS, 14.9% for MC40-25AS, 11.8% for MC40-25St, 18.9% for MC40-40AS, and 14.8% for MC45-40AS. The fractions of the other three species ranged from 0.2% to 6.5%. The residual fractions of Co in sludge after the bioleaching using MC were much less than those using a single bacterium. Moreover, since MC40-25As and MC40-25St presented high bioleaching efficiency for Co, it is necessary to evaluate the effect of the Ac and St ratios on bioleaching. The bioleaching BCR analysis results for Co in bioleached sludge using various Ac and St ratios in MC40 are shown in Fig. 6c. The leached fractions were 76.8% for MC40-50Ac, 73.9% for MC40-75Ac, 79.9% for MC40-50St and 73.3% for MC40-75St. The residual fractions were 16.0% for MC40-50Ac, 18.0% for MC40-75Ac, 13.8% for MC40-50St and 20.7% for MC40-75St. In addition, the BCR analysis of Co after bioleaching with various solid/liquid ratios of MC is displayed in Fig. 6d. As the solid/liquid ratio of MC40-25St increased from 10% to 50%, the leached fractions of Co declined from 82.9% to 58.9%, while the residual fractions rose continuously from 11.8% to 22.1%. Therefore, MC40-25St with a 10% solid/liquid ratio was the optimal adapted microbial consortium for bioleaching Co from bioleached sludge.

4. Conclusions

In summary, five microorganisms, including Af, Lf, Tt, Ac and St, and their six MC groups were applied to extract Ni and Co from the sludge from acid-leached waste lithium batteries. Among the single bacteria, Af presented the highest bioleaching ratio of 75.5% at 8 days for the extraction of Ni, and St showed the highest bioleaching ratio of 72.3% at 4 days for the extraction of Co. MC45-40AS could bioleach Ni efficiently with a ratio of 88.4% and an equilibrium time of 8 days. MC40-25St could bioleach Co efficiently with a ratio of 75.4% and an equilibrium time of 6 days. MC45-40AS and MC40-25St with a solid/liquid ratio of 10% were the optimal MC for the extraction of Ni and Co, respectively. The bioleaching ratios of MC45-40AS and MC40-25St were 1.17 times higher for Ni and 1.18 times higher for Co than that of the single bacterium. Moreover, from the BCR analysis results, MC45-40AS with a 10% solid/liquid ratio presented 90.3% for the leached fraction and 5.9% for the residual fraction for bioleaching Ni. MC40-25St with a 10% solid/liquid ratio exhibited 82.9% for the leached fraction and 11.8% for the residual fraction for bioleaching Co. This work paves the way for comprehending the migration and transformation of heavy metals from bioleached waste lithium batteries.

Author contributions

Haifei Lu: conceptualization, data curation, investigation, methodology, validation, writing-original draft, writing-review & editing. Houhu Zhang: writing-review & editing. Xiang Chen: data curation, writing-review & editing. Lili Wang: data curation, writing-review & editing. Xiaofei Yan: conceptualization, methodology, validation, supervision, writing-review & editing.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data that support the findings of this study are available on request from the corresponding author.

Supplementary information: Table S1. Concentrations of Ni in Ni slag (a) and Co in Co slag (b) measured by a handheld X-ray fluorescence spectrum. See DOI: https://doi.org/10.1039/d5ra07472j.

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