**Supplementary Information**

**Extraction of lithium from the simulated pyrometallurgical slag of spent lithium-ion batteries by binary eutectic molten carbonates**

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In general, the pyrometallurgical treatment of spent LIBs can be divided into three steps. In the first step (<300°C), the electrolyte is evaporated. In the second step (under 700°C), organics, such as plastic, are burned. Finally, in the third step (1200°C), reduction roasting transforms metals, such as Co, Ni, and Cu, into alloys. However, Li and Al cannot be reduced by carbon to form slag with the addition of slag-forming agents, such as CaO, SiO2, and Al2O3. Slag is usually composed of silica (30wt%–60wt%), calcium oxide (20wt%–35wt%), alumina (10wt%–25wt%), and lithium oxide (0.5wt%–15wt%) [1−2]. Actual slag also contains a small amount of metals, such as Fe and Ni, although ensuring that as little metal enters the slag as possible may be done during the disposal of spent LIBs [3]. Given these complexities, the composition of actual slag is difficult to analyze. Hence, simulated slag containing known main components (e.g., SiO2, CaO, Al2O3, and Li2O) was prepared for this study. The composition of the simulated slag and the roasting temperatures met the experimental ranges described above.

The particle size distribution and XRD pattern of the simulated slag are shown in Fig. S1 and S2, respectively.

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Fig. S1. Particle size distribution of the simulated slag.

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Fig. S2. XRD pattern of the simulated slag.

The effect of the mass of K2CO3 and Na2CO3 on the lithium extraction efficiency of the proposed system was studied, and the results are shown in Fig. S3. The lithium extraction efficiency increases with the increasing mass of the roasting agents.

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Fig. S3. Effect of the mass of K2CO3 or Na2CO3 on the lithium extraction efficiency of the proposed method (roasting conditions: temperature = 700℃ and time = 2 h; leaching conditions: water/roasted sample mass ratio = 10, temperature = 60℃, and time = 30 min).

The mass balance of the complete recycling process is shown in Table S1. The average standard reagent prices used in this study are presented in Table S2. The cost of slag is identical to that of cement because of its use as a building material. Roasting and leaching are the two key processes used to recover lithium from slag. The findings indicate that the recovery of slag is an economical endeavor, with an estimated profit margin of $429.92 per ton of slag.

Table S1. Mass balance of the complete recycling process mol

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Material | Input | Output (leaching solution) | Output (residue) | Error |
| Li+ | 1 | 0.94 | 0.04 | 0.02 |
| Al3+ | 1 | 0 | 0.97 | 0.03 |
| Ca2+ | 3.12 | 0 | 3.10 | 0.02 |
| SiO3− | 5.12 | 0 | 5.11 | 0.01 |
| K+ | 5.66 | 3.82 | 1.83 | 0.01 |
| Na+ | 13.21 | 11.42 | 1.77 | 0.02 |
| CO32− | 9.43 | 7.08 | 2.34 | 0.01 |

Table S2. Analysis of the economics of lithium recycling in this study

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Process | Reagents | | Unit price / ($⋅t−1) | Cost of reagent consumed in the process / $ | Price of recovered product / $ |
| Consumption | Recovery |
| Roasting | Slag |  | 188.99 | 188.99 |  |
| K2CO3 |  | 1070.3 | 834.84 |  |
| Na2CO3 |  | 214.16 | 299.82 |  |
| Energy consumption |  | 7.26 | 7.26 |  |
| Leaching | Deionized water |  | 22.34 | 692.54 |  |
|  | LiOH·H2O | 31019.98 |  | 2453.37 |
| Total cost |  |  |  | 2023.45 |  |
| Total recovery |  |  |  |  | 2453.37 |
| Profit in recycling | (2453.37 − 2023.45) = 429.92 | | | | |

**References**

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