

## **Supplementary Material**

### **Recycling Co and Li from spent lithium-ion batteries with the silicon of spent photovoltaics panels**

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## **List of Texts, Figures and Tables**

Text S1. Wafer pre-processing and economic analysis

Fig. S1. Composition of  $\text{LiCoO}_2$  cathode materials after pretreatment

Fig. S2. Silicon powder obtained after simple treatment

Fig. S3. Schematic diagram of co-processing of decommissioned PV panels to recover valuable elements from spent cathode materials.

Fig. S4. Total mass loss diagram before and after reaction

Fig. S5. SEM image of the sample after ball milling

Fig. S6. (a-b) SEM images of cathode materials after direct baking; (c-f) SEM images of added silicon mediated baking

Fig. S7. (a) Lithium leaching rate at different roasting times; (b) Lithium leaching rate under different leaching conditions

Fig. S8. (a) Cost analysis of processing 1kg of spent  $\text{LiCoO}_2$  cathode powder, (b) Carbon emission analysis

Table S1. Percentage of O before and after roasting

Table S2. Percentage of Co before and after roasting

Table S3. Recovery of valuable metals by combined thermal and wet processes

Table S4. Economic cost comparison (\$/kg capacity)

Table S5. Comparison of carbon emissions ( $\text{kg CO}_2/\text{kg treatment}$ )

#### Text S1. Wafer pre-processing and economic analysis

The obtained wafers were first reacted in 18 wt.% HCl solution for 15 min to dissolve Al. After washing and drying, they were reacted in 30 wt.% HNO<sub>3</sub> solution at 50 °C for 10 min to dissolve Ag, and then further washed and dried. The resulting silicon wafer was crushed into powder to obtain silicon powder as shown in Fig. S2. Before and after the reaction, the Al peak disappeared, removing the effects of Al and Ag, and the pretreated silicon powder was obtained. Some studies have confirmed that the method can almost completely dissolve Al and Ag<sup>1</sup>, and for the time being, the cost of the low concentration of HCl and HNO<sub>3</sub> used is low, and the amount of dissolved Al and Ag accounted for a relatively small percentage (3%), and the dissolving solution can be reused, but also the by-products of AgNO<sub>3</sub> and AlCl<sub>3</sub> for the benefit. The pretreatment process has low energy consumption and low cost.

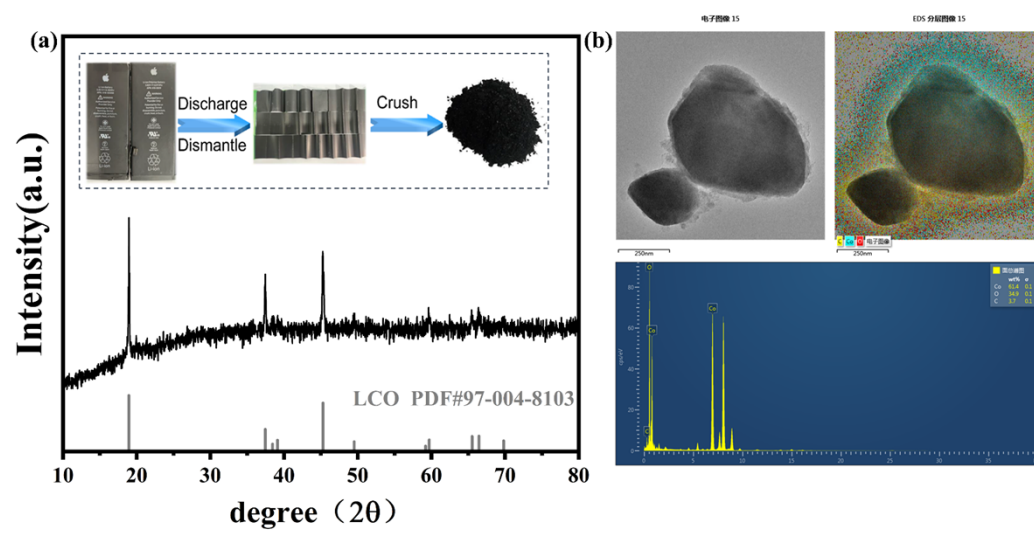


Fig. S1. Composition of  $\text{LiCoO}_2$  cathode materials after pretreatment

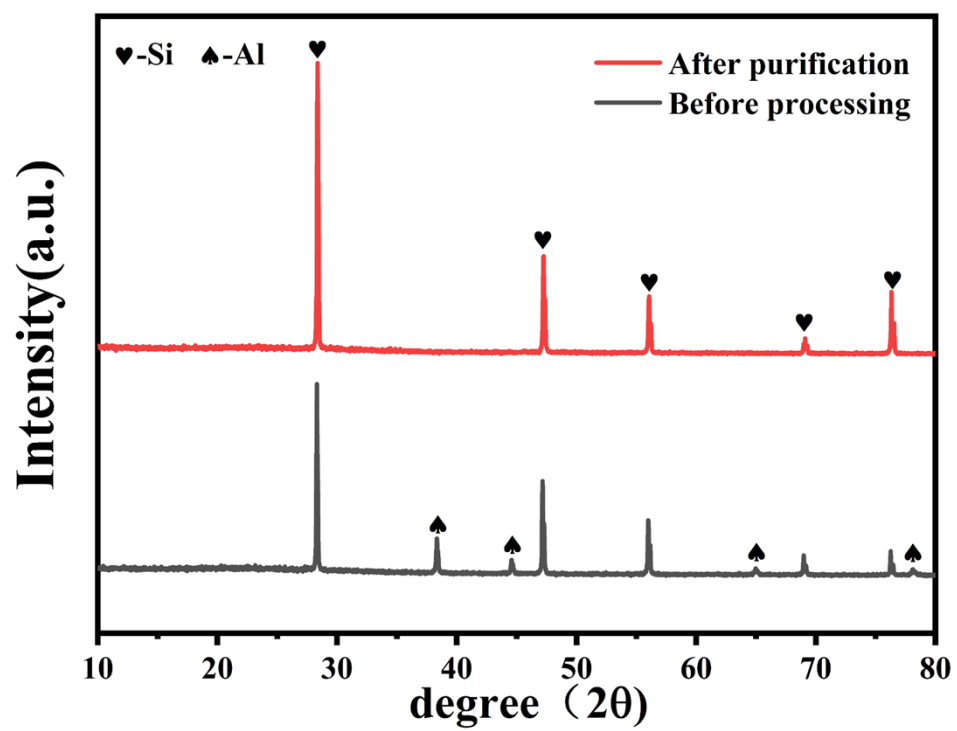


Fig. S2. Silicon powder obtained after simple treatment

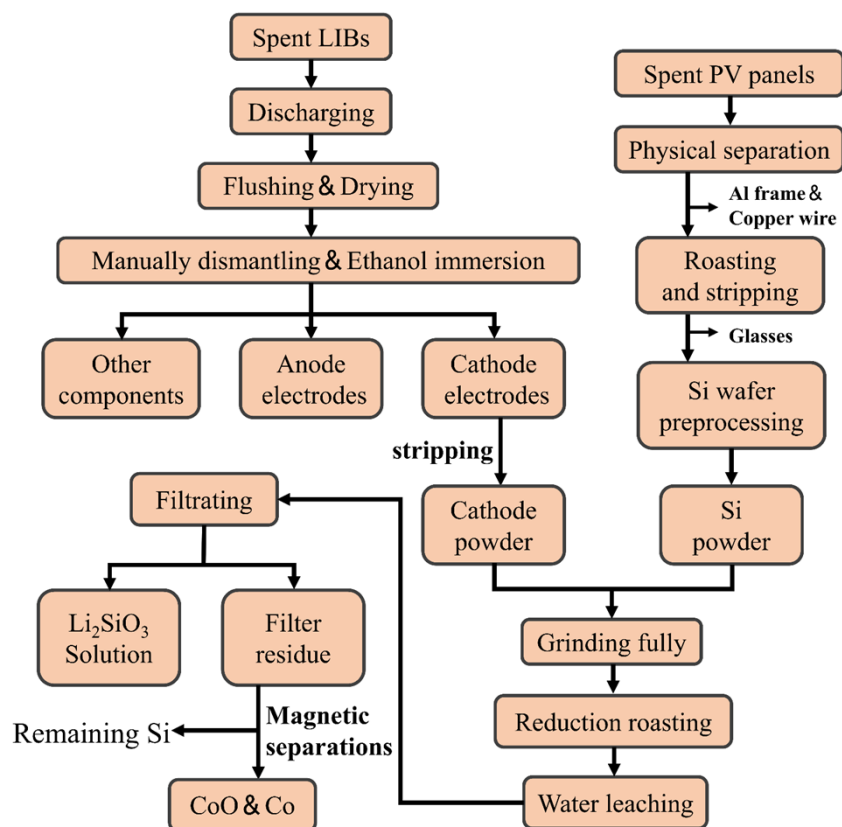


Fig.S3. Schematic diagram of co-processing of spent PV panels to recover valuable elements from waste cathode materials.

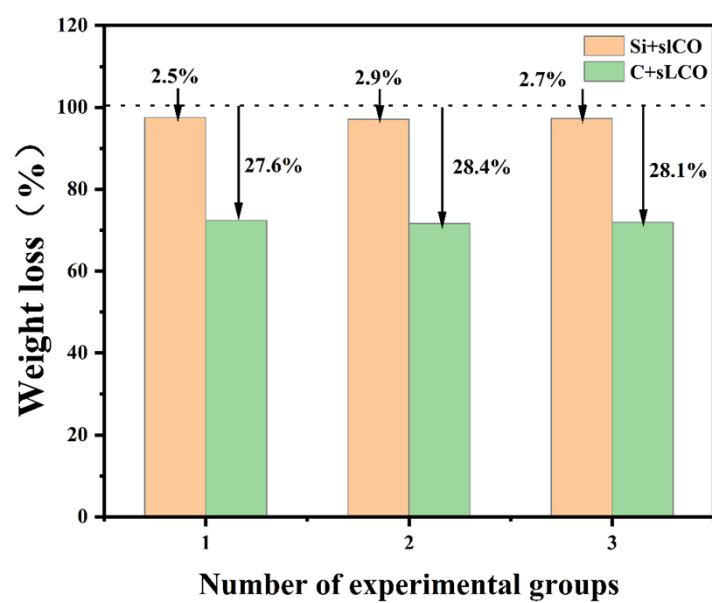


Fig. S4. Total mass loss diagram before and after reaction

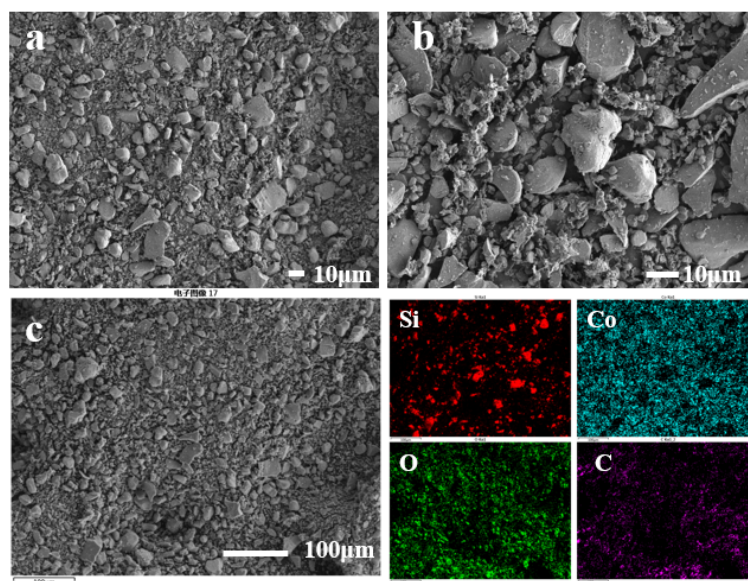


Fig. S5. SEM image of the sample after ball milling



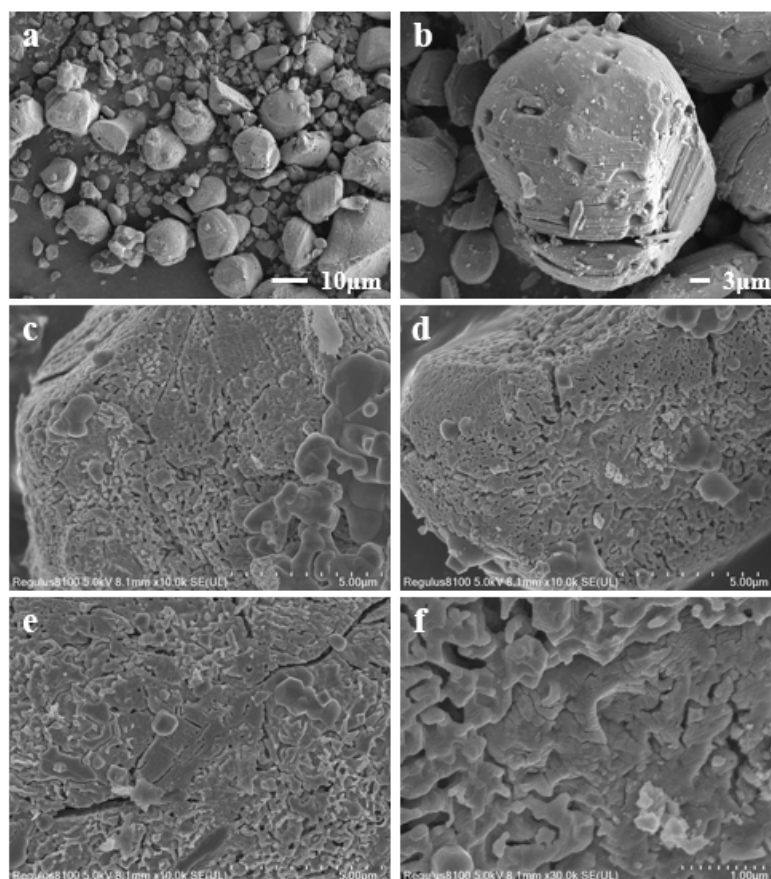


Fig. S6. (a-b) SEM images of cathode materials after direct baking; (c-f) SEM images of added silicon mediated baking

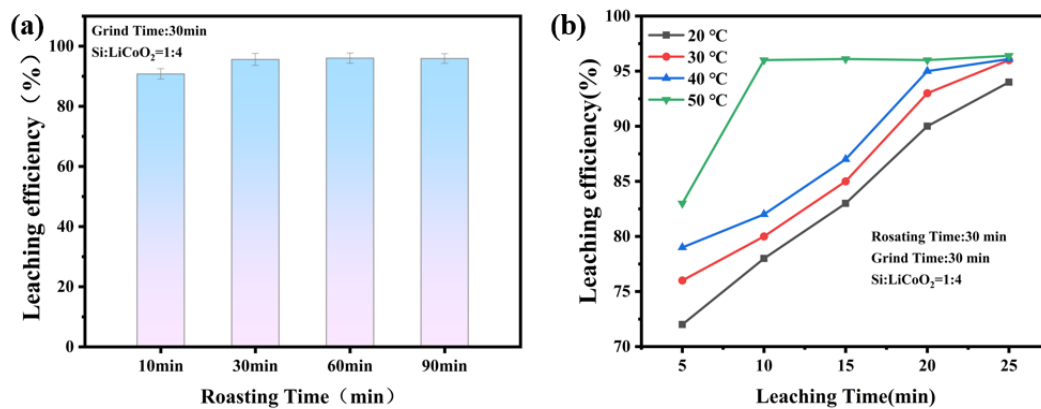


Fig. S7. (a) Lithium leaching rate at different roasting times; (b) Lithium leaching rate under different leaching condition

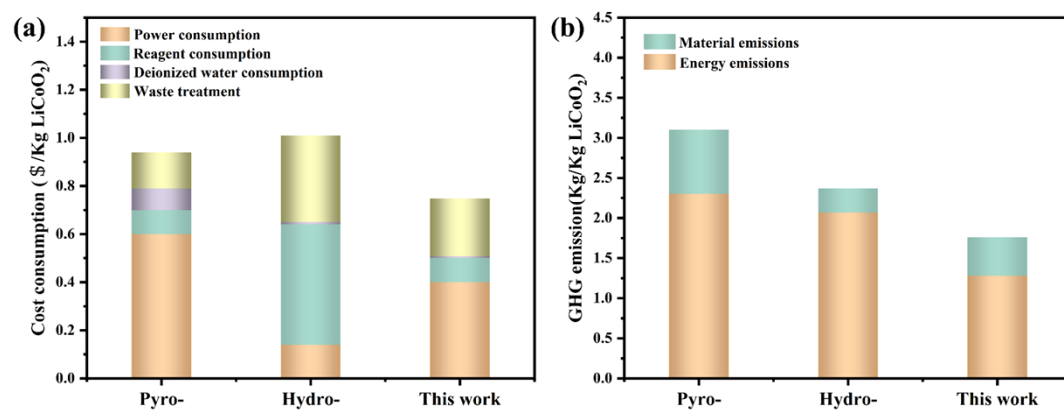


Fig. S8. (a) Cost analysis of processing 1kg of spent  $\text{LiCoO}_2$  cathode powder, (b) Carbon emission analysis

Table S1. Percentage of O before and after roasting

O	Lattice oxygen	Adsorbed oxygen	Other oxygen
Cathode	29.58%	68.24%	11.33%
Cathode-700 °C	50.88%	15.53%	39.59%
Cathode+Si-700 °C	19.54%	16.23%	49.07%

Table S2. Percentage of Co before and after roasting

Co	Co <sup>2+</sup>	Co <sup>3+</sup>
Cathode	35.89%	64.11%
Cathode-700 °C	40.79%	59.21%
Cathode+Si-700 °C	100%	0%

Table S3. Recovery of valuable metals by combined thermal and wet processes

Recovery	Leaching rate/%		Ref.
	Li	Co	
Biochar-assisted carbon reduction with water leaching	98	98.5	2
Microwave-assisted hydrogen reduction plus leaching	98.17	98.52	3
Diaphragm roasting plus leaching	93.2	-	4
Urea-assisted ammonium sulphate roasting plus leaching	94.94	95.47	5

Table S4. Economic cost comparison (\$/kg capacity)

Type of cost	Pyrometallurgy	Hydrometallurgy	This work
Power consumption	Electricity consumption: $2.5 \text{ kWh} \times 0.12 = 0.30$ Fuel consumption: $15 \text{ MJ} \times 0.02 = 0.30$	Electricity consumption: $1.2 \text{ kWh} \times 0.12 =$ 0.14	Electricity consumption: $1.5 \text{ kWh} \times 0.12 = 0.18$ Fuel consumption: $10 \text{ MJ} \times 0.02 = 0.20$
Reagent consumption	Acid consumption $0.2 \text{ kg} \times 0.5 = 0.10$	Acid consumption $1.0 \text{ kg} \times 0.5 = 0.50$	Recycled silicon $0.2 \text{ kg} \times 0.5 = 0.10$
Deionized water consumption	$9 \text{ L} \times 0.001 = 0.09$	$10 \text{ L} \times 0.001 =$ 0.01	$8 \text{ L} \times 0.001 = 0.008$
Waste treatment	$0.5 \text{ kg} \times 0.3 = 0.15$	$1.2 \text{ kg} \times 0.3 = 0.36$	$0.8 \text{ kg} \times 0.3 = 0.24$
Total cost (\$/kg)	0.94	1.01	0.73

Table S5. Comparison of carbon emissions (kg CO<sub>2</sub>/kg treatment)

Sources of emissions	Pyrometallurgy	Hydrometallurgy	This work
Energy emissions	High temperature consumption	Stirring, extraction $1.2 \text{ kWh} \times 0.475 = 0.57$	Medium temperature consumption
	$2.5 \text{ kWh} \times 0.475 = 1.19$	Acid consumption (H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub> )	$1.5 \text{ kWh} \times 0.475 = 0.71$
	Fuel consumption $15 \text{ MJ} \times 0.074 = 1.11$	$1.5 \text{ kg} \times 1 = 1.5$	Fuel consumption $10 \text{ MJ} \times 0.057 = 0.57$
Material emissions	Direct process emissions (CO <sub>2</sub> ) 0.8	Chemical precipitation & extractant emissions 0.3	Silicon recovery process 0.48
Total carbon emissions (kg CO <sub>2</sub> /kg)	3.1	2.37	1.76



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