Supporting Information

Directional conversion of valuable components from spent carbon cathode via high-temperature roasting

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Table of contents

Table S1 ..... S3

Table S2 ..... S4

Table S3 ..... S5

Table S4 ..... S5

Table S5 ..... S6

Table S6 ..... S7

Fig. S1 ..... S8

Fig. S2 ..... S9

Fig. S3 ..... S9

Fig. S4 ..... S10

Fig. S5 ..... S10

Table S1

Experimental scheme for optimization of synthetic cryolite parameters.

Fig.	Fluoride ion concentration/mg·L^{-1}	Inducer/g	pH	Sustain temperature/℃	Holding time/h
9(a) - (c)	500 - 2500	0.2	9	70	24
9(d) - (f)	2000	0.1 - 0.3	9	70	24
9(g) - (i)	2000	0.2	5 - 9	70	24

Table S2

Possible reactions of Na, Al, and F ions in solution.

Eqs.	Reaction	Log K
6	$H^+ + OH^- \rightleftharpoons H_2O$	13.997
7	$H^+ + F^- \rightleftharpoons HF$	3.17
8	$HF + F^- \rightleftharpoons HF_2^-$	0.5
9	$Na^+ + F^- \rightleftharpoons NaF\ (aq)$	0.02
10	$Al^{3+} + H_2O \rightleftharpoons Al(OH)^{2+} + H^+$	-5.00
11	$Al^{3+} + 2H_2O \rightleftharpoons Al(OH)_2^+ + 2H^+$	-10.1
12	$Al^{3+} + 3H_2O \rightleftharpoons Al(OH)_3 + 3H^+$	-16.8
13	$Al^{3+} + 4H_2O \rightleftharpoons Al(OH)_4^- + 4H^+$	-23
14	$Al^{3+} + F^- + H_2O \rightleftharpoons AlF(OH)^+ + H^+$	1.4
15	$Al^{3+} + F^- + 2H_2O \rightleftharpoons AlF(OH)_2 + 2H^+$	-4.9
16	$Al^{3+} + F^- + 3H_2O \rightleftharpoons AlF(OH)_3^- + 3H^+$	-11.9
17	$Al^{3+} + 2F^- + H_2O \rightleftharpoons AlF_2(OH) + H^+$	6.1
18	$Al^{3+} + 2F^- + 2H_2O \rightleftharpoons AlF_2(OH)_2^- + 2H^+$	-0.8
19	$Al^{3+} + 3F^- + H_2O \rightleftharpoons AlF_3(OH)^- + H^+$	9.6
20	$Al^{3+} + F^- \rightleftharpoons AlF^{2+}$	7
21	$Al^{3+} + 2F^- \rightleftharpoons AlF_2^+$	12.7
22	$Al^{3+} + 3F^- \rightleftharpoons AlF_3$	16.8
23	$Al^{3+} + 4F^- \rightleftharpoons AlF_4^-$	19.4
24	$Al^{3+} + 5F^- \rightleftharpoons AlF_5^{2-}$	20.6
25	$Al^{3+} + 6F^- \rightleftharpoons AlF_6^{3-}$	26.5
26	$3Na^+ + AlF_6^{3-} \rightleftharpoons Na_3AlF_6\ (s)$	9.375

Table S3

The material and ion balances for F, Na, and Al.

Eqs.	Reaction
27	$[F]_T=[F^-]+[HF]+2\times[HF_2^-]+[NaF(aq)]+[AlF(OH)^+]+[AlF(OH)_2]+[AlF(OH)_3^-]+2\times[AlF_2(OH)]+2\times[AlF_2(OH)_2^-]+3\times[AlF_3(OH)^-]+[AlF^{2+}]+2\times[AlF_2^+]+3\times[AlF_3]+4\times[AlF_4^-]+5\times[AlF_5^{2-}]+6\times[AlF_6^{3-}]$
28	$[Na]_T=[Na^+]+[NaF(aq)]$
29	$[Al]_T=[Al^{3+}]+[Al(OH)^{2+}]+[Al(OH)_2^+]+[Al(OH)_3]+[Al(OH)_4^-]+[AlF(OH)^+]+[AlF(OH)_2]+[AlF(OH)_3^-]+[AlF_2(OH)]+[AlF_2(OH)_2^-]+[AlF_3(OH)^-]+[AlF^{2+}]+[AlF_2^+]+[AlF_3]+[AlF_4^-]+[AlF_5^{2-}]+[AlF_6^{3-}]$
30	$[F^-]+[HF_2^-]+[Al(OH)_4^-]+[AlF(OH)_3^-]+[AlF_2(OH)_2^-]+[AlF_3(OH)^-]+[AlF_4^-]+2\times[AlF_5^{2-}]+3\times[AlF_6^{3-}]+[OH^-]+2\times[CO_3^{2-}]=3\times[Al^{3+}]+2\times[Al(OH)^{2+}]+[Al(OH)_2^+]+[AlF(OH)^+]+2\times[AlF^{2+}]+[AlF_2^+]+[H^+]+[Na^+]$

The economic evaluation of 1kg spent cathode carbon(SCC) was carried out according to the experimental process.

Table S4

Price of electricity, water, and chemical reagents.

No.	Substance	Price
1	Electricity	\$0.07/kW·h
2	Water	\$0.0006/L
3	Aluminium-based inducer	\$0.12/kg
4	Acid leaching reagent	\$0.1/L
5	High-purity graphite	\$0.58/kg
6	Na_3AlF_6	\$1.59/kg

The detailed calculation is as follows:

Reagent and water cost:

Water**:**40L×\$0.0006/L=\$0.024

Aluminium-based inducer**:**0.08kg×\$0.12/kg=\$0.0096

Acid leaching reagent**:**0.5L×\$0.1/L=\$0.05

Energy Costs:

Bake oven:2 h×0.8 kW×$0.07/kW・h=$0.112 (calculated based on 2 hour of drying)

High temperature tubular furnace**:**8 kWh×$0.07/kW·h=$0.56

Water-bath**:**0.5kW·h×$0.07/kW·h=$0.035

Product revenue:

High-purity graphite:0.75kg×$0.58/kg=$0.435

Na_3AlF_6:0.15kg×$1.59/kg=$0.2385

Table S5

Economic benefit analysis.

Item	Cost expenditure($)	Recovery value($)	Economic benefit($)
Aluminium-based inducer	-0.0096	0	-0.0096
Acid leaching reagent	-0.05	0	-0.05
Water	-0.024	0	-0.024
Bake oven	-0.112	0	-0.112
High temperature tubular furnace	-0.56	0	-0.56
Water-bath	-0.035	0	-0.035
High-purity graphite	0	0.318	0.318
Na_3AlF_6	0	0.2385	0.2385
Total	-0.7906	0.5565	-0.2341

The table shows that the final cost for processing 1kg of SCC is $0.2341, lower than the $0.5714/kg cost of traditional landfill disposal.

Table S6

Comparison of economic efficiency with mature technologies (microwave, vacuum roasting, molten salt)

Technology type	Capital cost of equipment	Cost of energy consumption	Maintenance cost	Product quality stability	Comprehensive economic benefits
Atmospheric furnace roasting	Middle,the cost of purchasing the equipment is moderate.	High,high temperature (1500-1600 ℃) results in high energy consumption.	Middle,regular maintenance of equipment components	High,simple process control	Middle,the initial investment is relatively large, but long-term returns can be seen later.
Microwave roasting	High,the equipment cost is relatively high.	Low,high microwave heating efficiency	High,equipment is difficult to maintaine	High,uniform heating ensures stable product quality	Low,low energy cost, high product quality, suitable for high value-added products
Vacuum roasting	High, vacuum equipment and sealing system costs are elevated	Middle,the energy consumption in vacuum environment is relatively low.	High,high maintenance requirements for vacuum systems	High,prevent oxidation, high product purity	Low,suitable for products with high purity requirements, but with higher equipment and maintenance costs
Molten salt roasting	Middle,the cost of molten salt equipment and molten salt medium is moderate	Middle,molten salt heat transfer efficiency is higher	Middle,molten salt requires regular replacement and treatment	Low,affected by the properties of molten salt and process control	High,the overall cost and benefit are relatively balanced

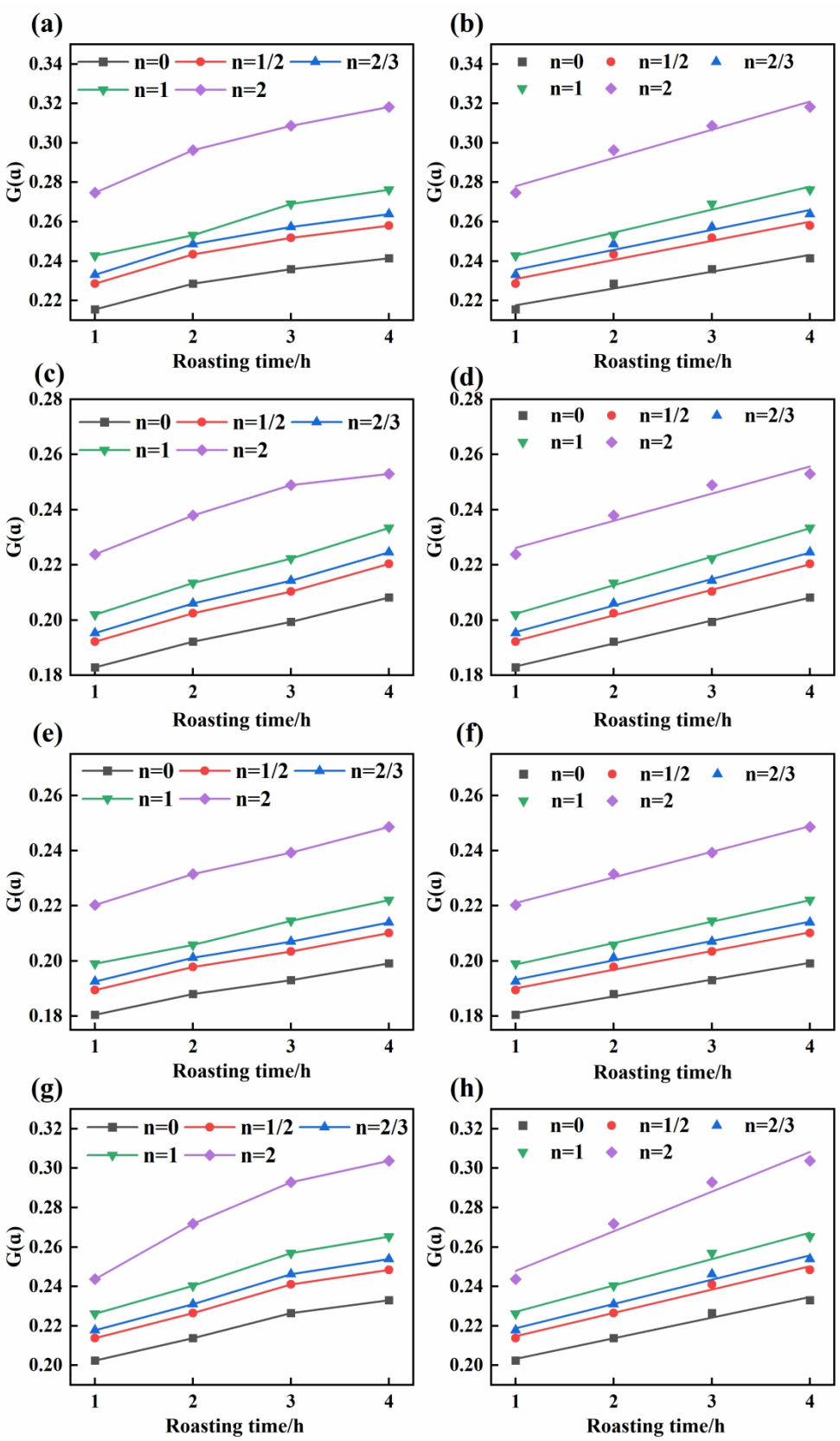


Figure S1. Relationship curves and linear fitting plots of G(α) versus t at 1400 ℃: (a)-(b) 5-3 mm, (c)-(d) 3-1 mm, (e)-(f) 1-0.15 mm, (g)-(h) <0.15 mm.

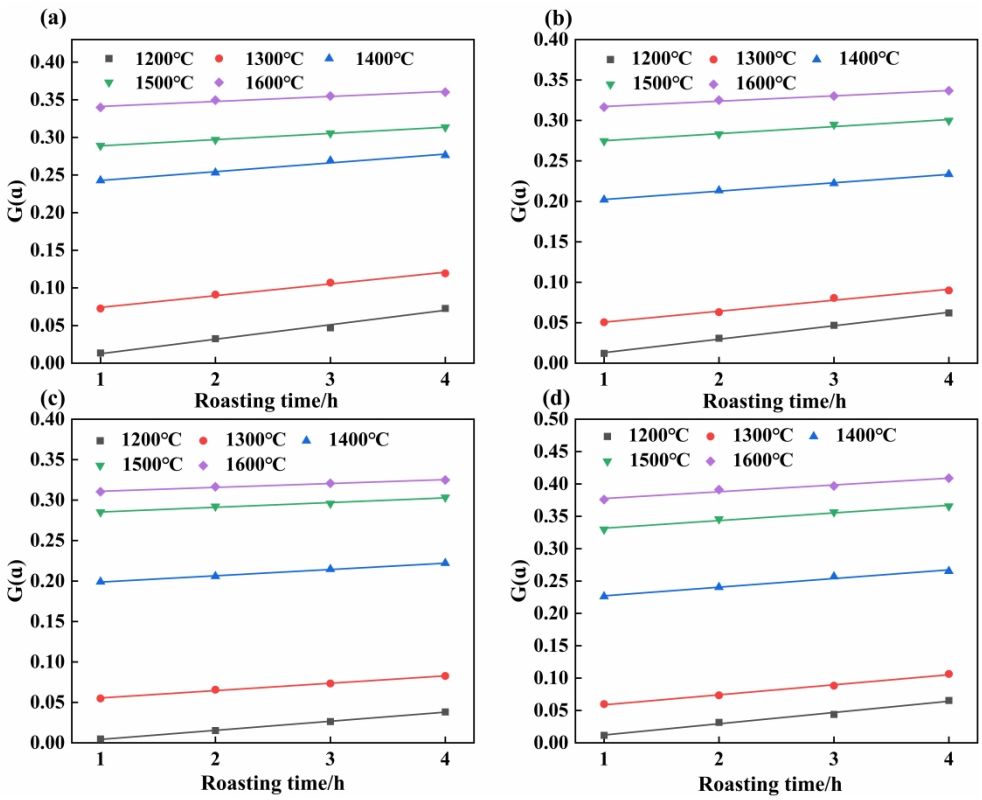


Figure S2. Linear fitting plots of G(α) versus t at different roasting temperatures: (a) 5-3 mm, (b) 3-1 mm, (c) 1-0.15 mm, (d) <0.15 mm.

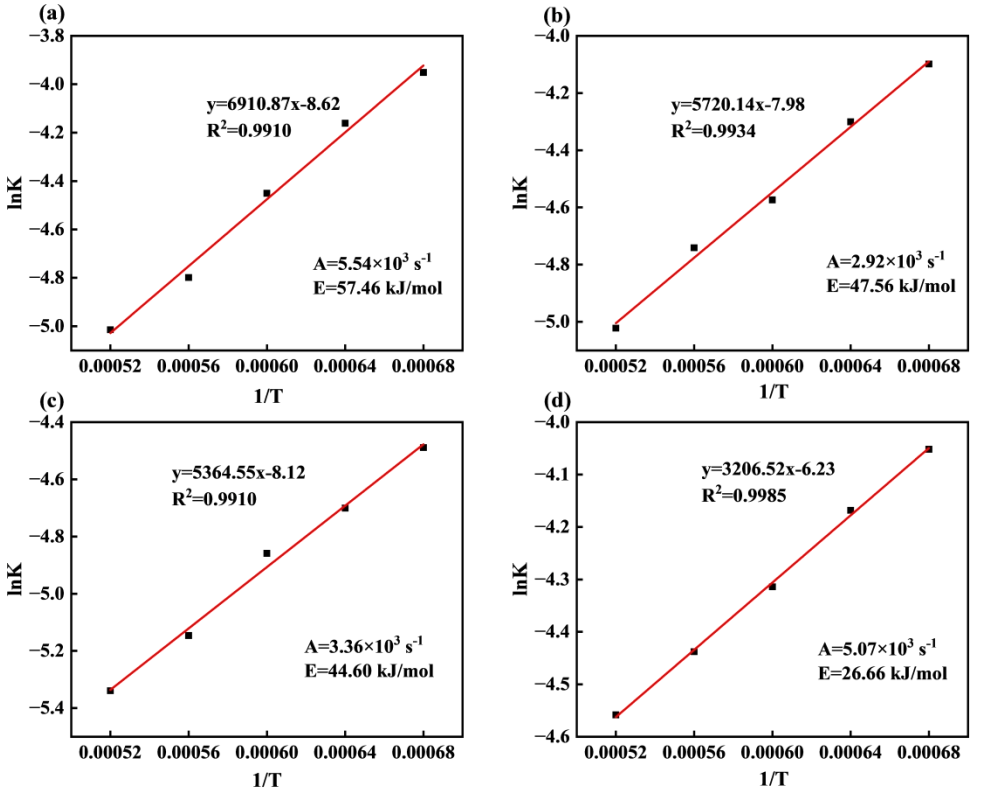


Figure S3. Linear fitting plots of lnK versus 1/T for different particle sizes: (a) 5-3 mm, (b) 3-1 mm, (c) 1-0.15 mm, (d) <0.15 mm.

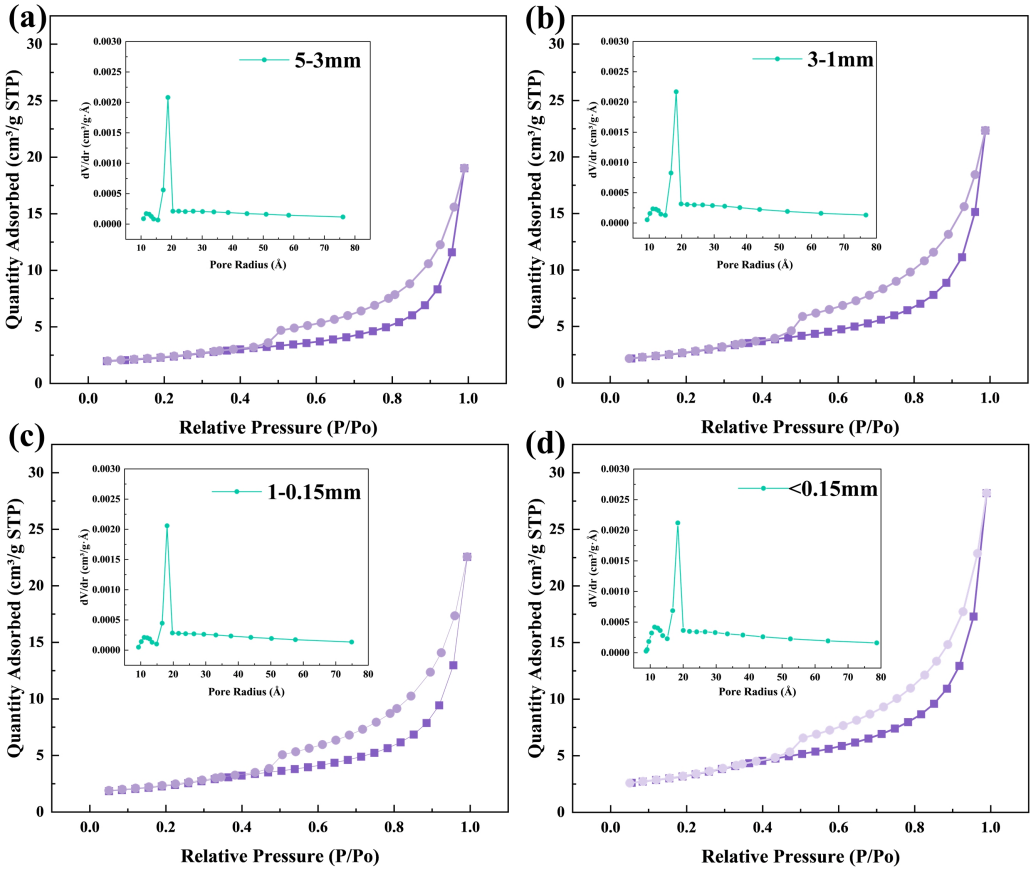


Figure S4. RG adsorption-desorption curves and pore size distribution: (a)5-3 mm，(b)3-1 mm，(c)1-0.15 mm，(d)-0.15 mm.

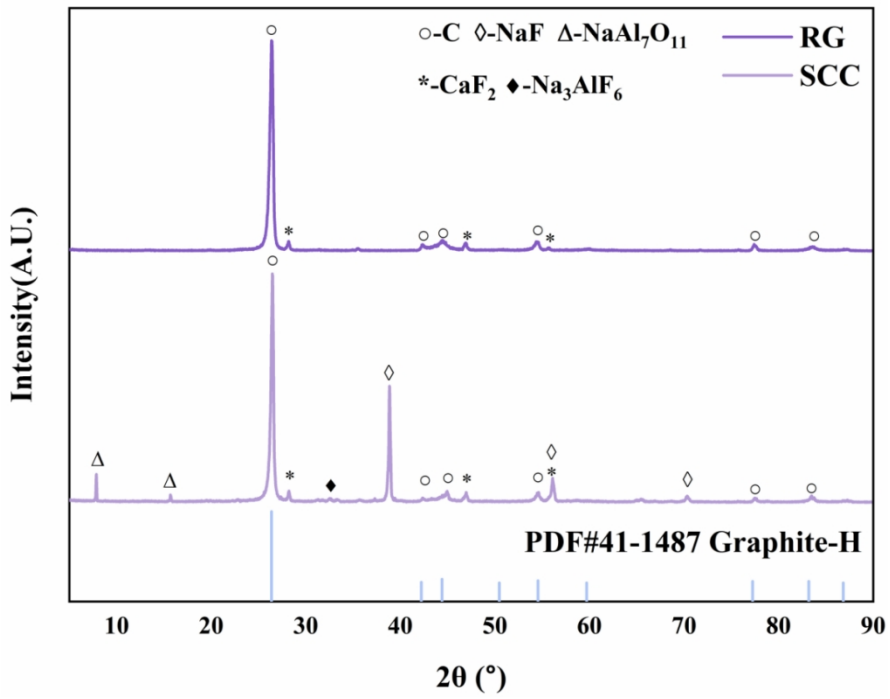


Figure S5. XRD of SCC and RG