Electronic supplementary information (ESI)

A new method for the preparation of high-purity CO_2-absorbing Li_3NaSiO_4 powder using lithium silicate and sodium carbonate

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Preparation of Li_2SiO_3

Nominal amounts of Li_2CO_3 powder (99.9 %, Furuuchi Chemical Co.) and SiO_2 powder (99.9%, Kojundo Chemical Laboratory Co., Ltd) were mixed in ethanol using an Al_2O_3 mortar and pestle. The mixed powder was heated at 950 °C for 24 h in air, followed by pulverization in the Al_2O_3 mortar and pestle. The obtained powder was subsequently heated at 1000 °C for 24 h in air two times.

Fig. S1 shows the XRD pattern of the obtained powder. All the peaks were successfully indexed to orthorhombic symmetry with a = 5.398 Å, b = 9.387 Å, and c = 4.661 Å, which confirmed the synthesis of single-phase Li_2SiO_3 (JCPDS No. 29-0829).

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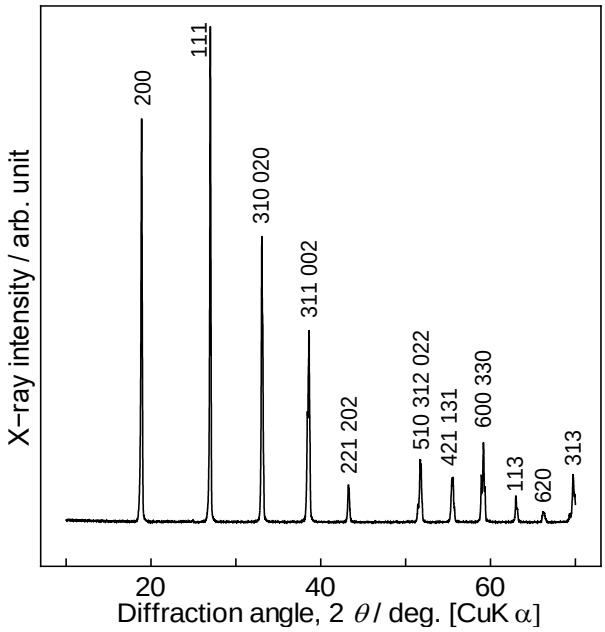

Fig. S1 XRD pattern of the Li_2SiO_3 compound employed in this study.

Preparation of Li_4SiO_4

Nominal amounts of Li_2CO_3 powder (99.9 %, Furuuchi Chemical Co.) and SiO_2 powder (99.9%, Furuuchi Chemical Co.) were mixed in ethanol using the Al_2O_3 mortar and pestle. The mixed powder was heated at 700 °C for 12 h in air.

Fig. S2 shows the XRD pattern of the obtained powder. All the peaks were successfully indexed to monoclinic symmetry with a = 5.298 Å, b = 6.102 Å, c = 5.150 Å, and β = 90.25°, which validated the synthesis of single-phase Li_4SiO_4 (JCPDS No. 37-1472).

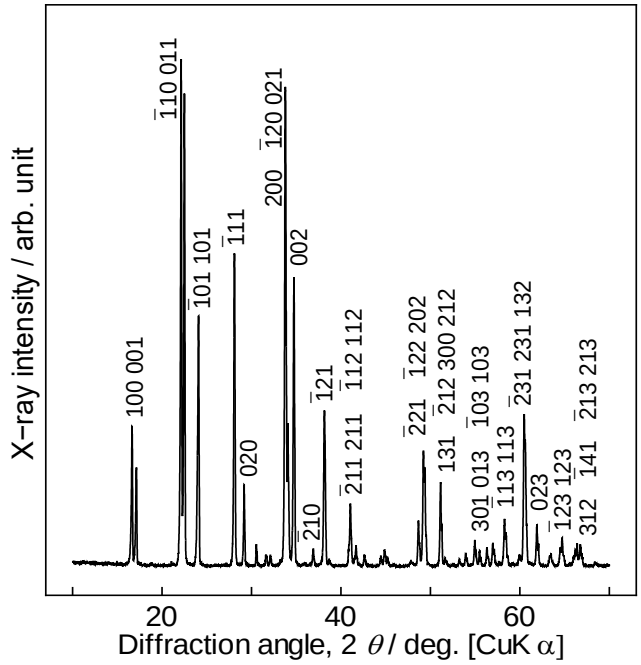

Fig. S2 XRD pattern of the Li_4SiO_4 compound employed in this study.

Preparation of $LiNaCO_3$

Nominal amounts of Li_2CO_3 powder (99.9 %, Furuuchi Chemical Co.) and Na_2CO_3 powder (99.9%, FUJIFILM WAKO Chem.) were mixed in ethanol using the Al_2O_3 mortar and pestle. The powder was heated at 500 °C for 24 h in air.

Fig. S3 shows the XRD pattern of the obtained powder. All the peaks were successfully indexed to the C2/m (No. 12) space group with a = 14.285 Å, b = 24.750 Å, c = 3.302 Å, and β = 91.7º, thereby confirming the preparation of single-phase $LiNaCO_3$ (JCPDS No. 34-1193).

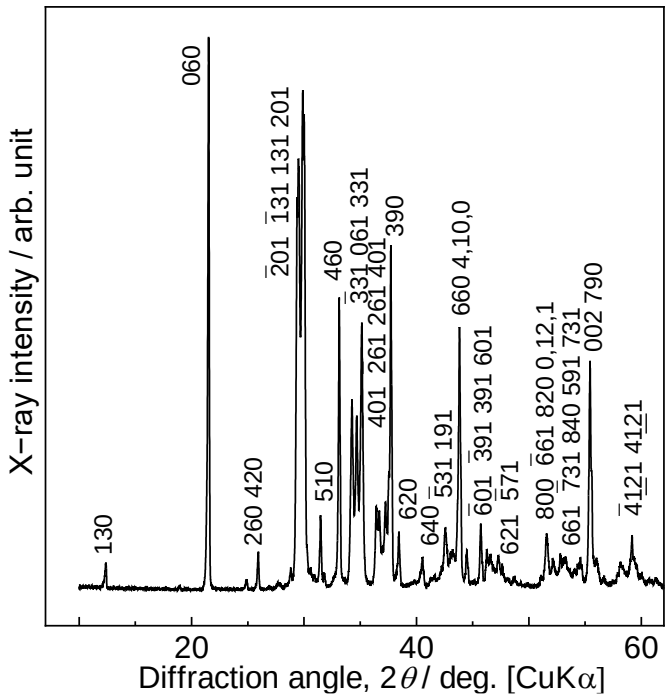

Fig. S3 XRD pattern of the $LiNaCO_3$ compound used in this study.

Morphology of powder specimens after CO_2 absorption

Fig. S4 shows the SEM images of the powder after CO_2 absorption. CO_2 absorption was performed at 700 ºC and followed by quenching to 25 ºC under CO_2 gas flow in a TG-DTA apparatus. The initial Li_3NaSiO_4 powder was prepared from (a) $Li_4SiO_4/Li_2SiO_3/SiO_2$ and (b) $Li_2CO_3/Na_2CO_3/SiO_2$. XRD measurements revealed that both specimens were a mixture of $LiNaCO_3$ and Li_2SiO_3.[1]

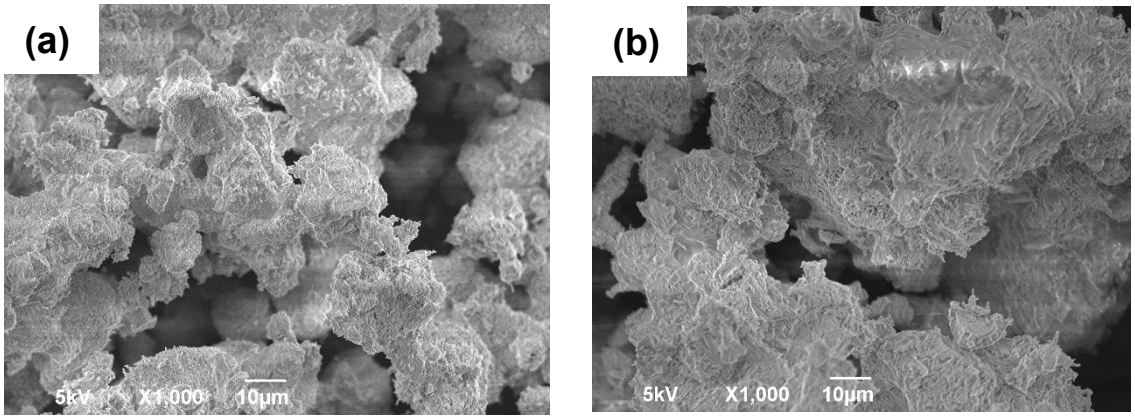

Fig. S4 SEM images of the powder after CO_2 absorption at 700 ºC. Initial Li_3NaSiO_4 powder was prepared from (a) $Li_4SiO_4/Li_2SiO_3/SiO_2$ and (b) $Li_2CO_3/Na_2CO_3/SiO_2$.

Less difference in morphology between the two powder specimens was observed compared to the initial Li_3NaSiO_4 powders depicted in Fig. 8. At 700 ºC in CO_2 atmosphere, Li_3NaSiO_4 decomposes to solid Li_2SiO_3 powder and liquid $LiNaCO_3$. It was prospected that the size and shape of Li_2SiO_3 particles generated by the decomposition was similar between the two kinds of powder, resulting in similar morphology of the powder mixture of Li_2SiO_3 and $LiNaCO_3$, which solidified from the liquid phase during cooling at approximately 500 ºC.

Reference

1. M. Hirai, E. Niwa and T. Hashimoto, *Dalton Trans.*, 2021, **50**, 5301–5310