Electronic supplementary information

A green and scalable synthesis of highly stable Ca-based sorbents for CO_2 capture

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Table S1 Chemical compositions of the sorbents

Samples	CaO (%)	SiO_2 (%)	Al_2O_3 (%)	Fe_2O_3 (%)	K_2O (%)	TiO_2 (%)	MgO (%)	Trace (%)
FA	2.80	53.96	24.04	11.32	3.70	1.37	1.10	1.71
CaO [a]	97.90	0.93	0.50	0.45	0.03	0.07	0.01	0.11
CaO-FA [b]	89.88	5.98	2.19	1.13	0.13	0.16	0.18	0.35

[a] Including the sorbents of "CaO-0h" and "CaO-2h"; [b] including the sorbents of "CaO-FA-0h", "CaO-FA-2h", "CaO-FA-6h", and "CaO-FA-12h".

Table S2 Tammann temperatures (T_T) of the typical minerals

Samples	Melting temperature (ºC)	T_T [a] (ºC)	Samples	Melting temperature (ºC)	T_T (ºC)
CaO [1]	2613	1170	$CaCO_3$ [2]	1320 [b]	524
Al_2O_3 [1]	2054	891	$Ca_2Al_2SiO_7$ [3]	1593	797
MgO [1]	2825	1276	$Ca_{12}Al_{14}O_{33}$ [4]	1722	725
SiO_2 [1]	1722	725	$Al_{4.8}Si_{1.2}O_{9.6}$	1910	955
TiO_2 [1]	1843	785	Ca_2SiO_4 [5]	2130	929
Y_2O_3 [1]	2439	1083	$CaTiO_3$ [5]	1975	851
ZrO_2 [1]	2710	1219	$CaZrO_3$ [5]	2345	1036

[a] $T_T \approx 0.5\ T_{melting}$, referring to the temperatures at which sintering occurs. [b] Determined at 100 MPa.

1 CRC handbook of chemistry and physics 95th edition internet version 2014-2015, http://www.hbcpnetbase.com/.
2 P. J. Wyllie and O. F. Tuttle, *J. Petrol.*, 1960, **1**, 1-46.
3 G. Gruener, K. Dembinski, A. Bouvier, J.P. Loup and P. Odier, *Eur. Phys. J.-Appl. Phys.*, 1998, **4**, 101-106.
4 J. E. Medvedeva, E. N. Teasley, M. D. Hoffman, *Phys. Rev. B*, 2007, **76**, 155107.
5 M. Zhao, J. Shi, X. Zhong, S. C. Tian, J. Blamey, J. G. Jiang, P. S. Fennell, *Energy Environ. Sci.*, 2014, **7**, 3291-3295.

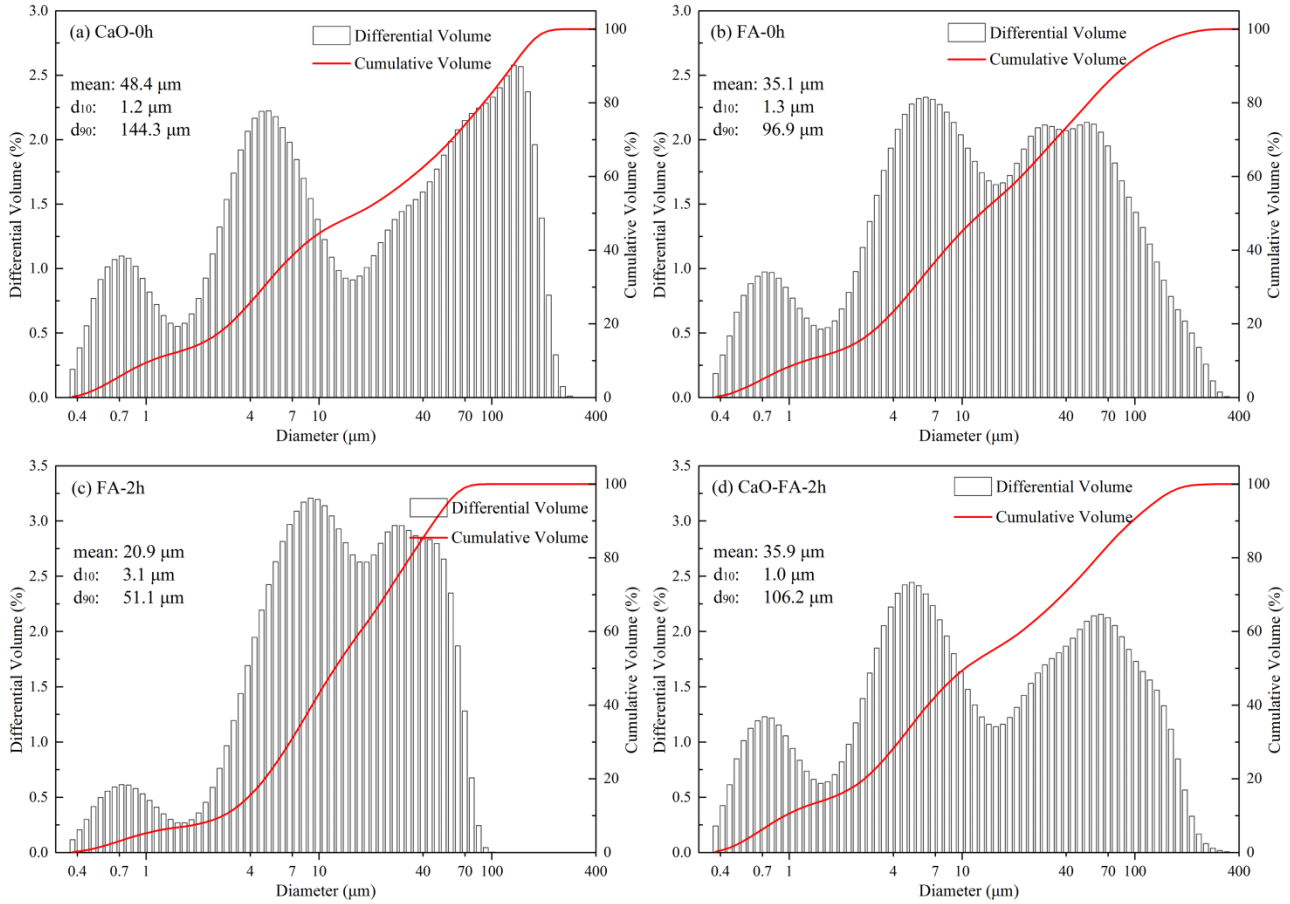

Fig. S1 Particle size distribution curves of samples: (a) "CaO-0h"; (b) "FA-0h"; (c) "FA-2h"; (d) "CaO-FA-2h".

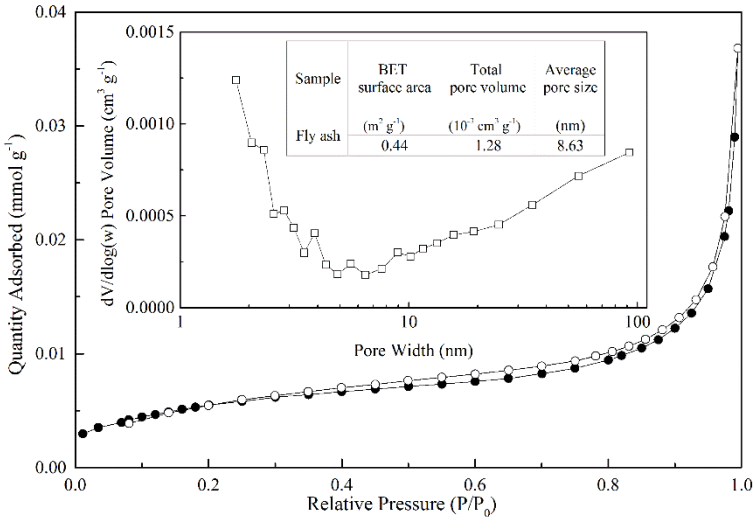

Fig. S2 N_2 physisorption isotherms and pore volume distributions (inset) of the fly ash.

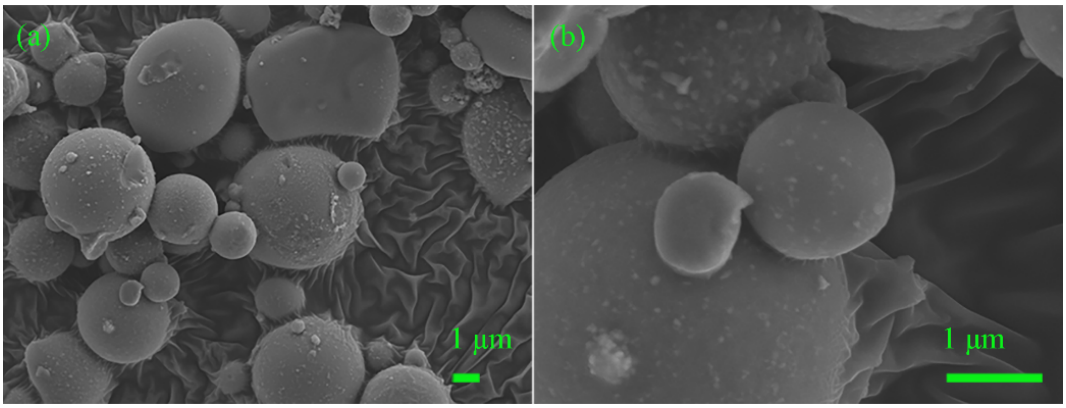

Fig. S3 SEM images of the fly ash: (a) ×5,000; (b) ×20,000.

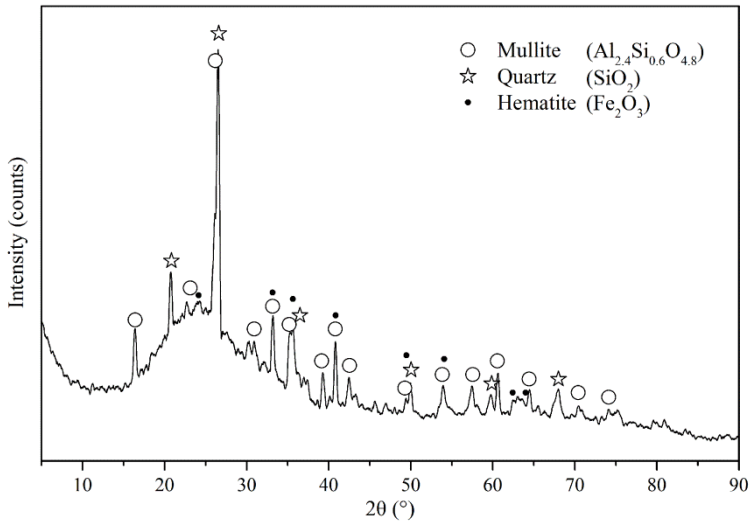

Fig. S4 XRD patterns of the fly ash.

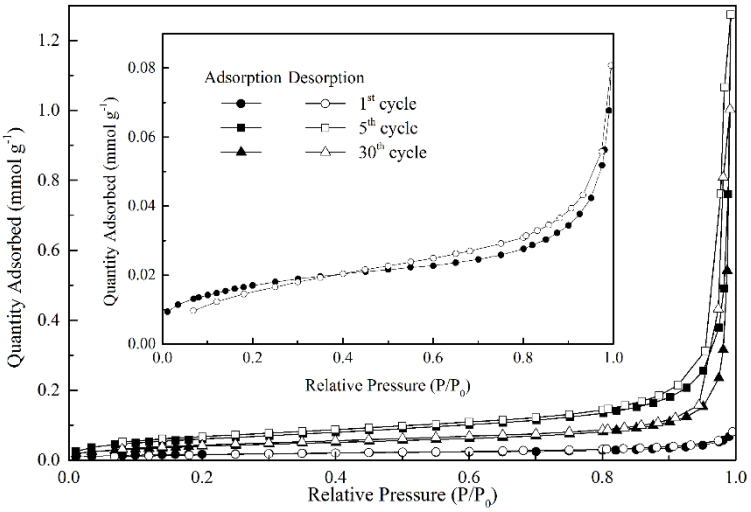

Fig. S5 N_2 physisorption isotherms of "CaO-2h" before the 1st, 5th, and 30th cycle, separately.

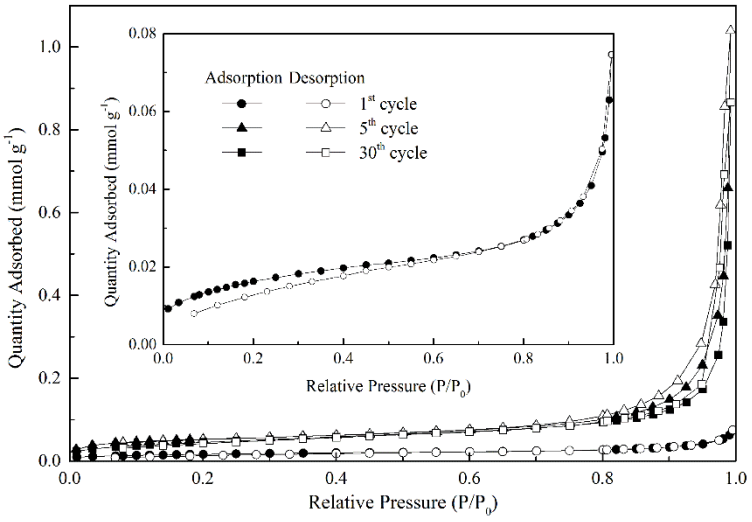

Fig. S6 N_2 physisorption isotherms of "CaO-FA-2h" before the 1st, 5th, and 30th cycle, separately.

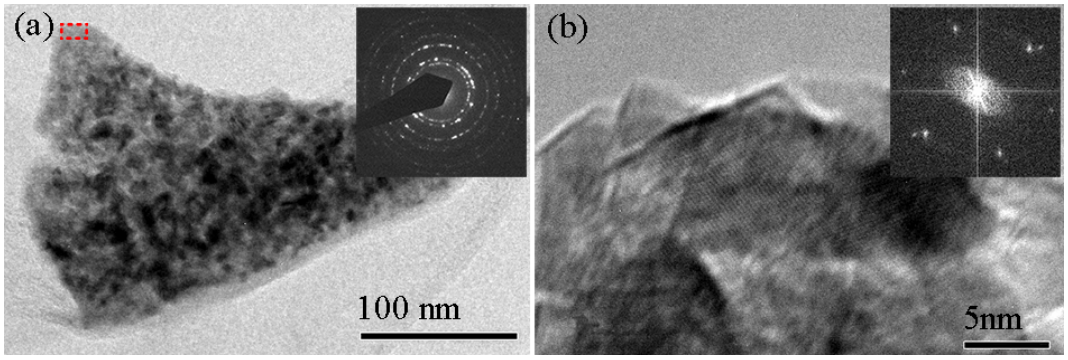

Fig. S7 (a) TEM image and electron diffraction pattern of "CaO-2h"; (b) HRTEM image and electron diffraction pattern of the selected area in (a)**.**

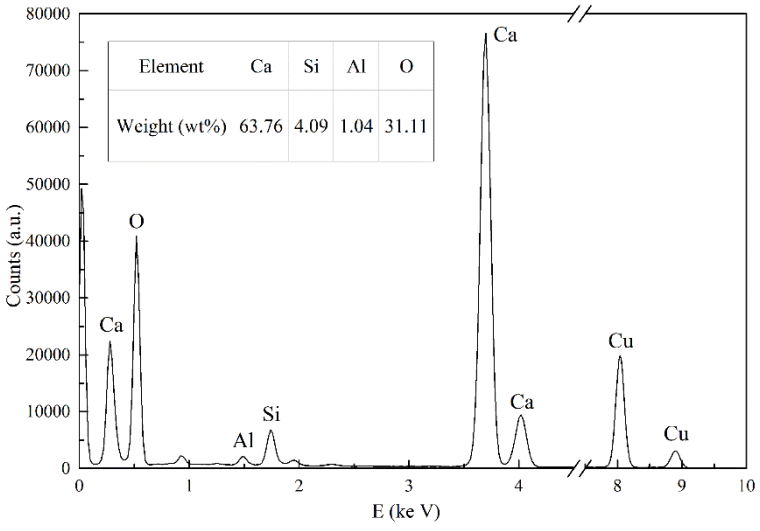

Fig. S8 EDX spectrum averaged on the region of "CaO-FA-2h" (Fig. 2-(d)). A quantitative mapping analysis yields a wt % of 63.76% for Ca, 4.09% for Si, 1.04% for Al and 31.11% for O.

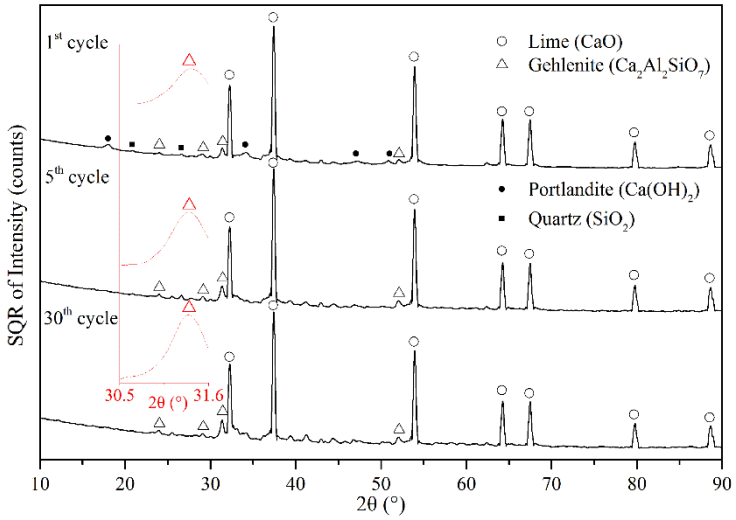

Fig. S9 XRD patterns of "CaO-FA-2h" before the 1st, 5th, and 30th cycle, separately.