Supporting information

Evaluation and mechanism analysis of alkaline earth metal CaO for Cd solidification enhancement in lightweight aggregates preparation

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1. Characterizations

The macroscopic properties of the lightweight aggregate sample tested in this experiment include volume expansion rate, apparent density, water absorption rate, and single particle compressive strength. Before testing, it is necessary to ensure that the sample is in a dry state. Among them, the volume expansion rate is characterized by the expansion index (BI), and the apparent density (ρ). Besides, the determination of water absorption (W) was carried out in accordance with the national standard GB/T 17431.2-2010, and the single particle compressive strength (P) was obtained by testing with a laboratory fully automatic cement bending and compression testing machine. The final values of all test items are the average of six lightweight aggregate samples. The corresponding calculation formula of each test is as follows:

Bloating index (BI)：$$BI = \frac{V_2 - V_1}{V_1} \times 100\%$$

Apparent density (ρ)：$$\rho = M_1/V_2$$

Water absorption (W)：$$W = \frac{M_2}{M_1} \times 100\%$$

Compressive strength (P)：$$P = 2.8F_c/(\pi X^2)$$

V_1 and V_2 represent the sample volumes before and after sintering of the LWAs, M_1 and M_2 represent the sample masses before and after water absorption of the LWAs, Fc (kN) is the pressure value (fracture load) at which the LWAs sample fracture failure under external force, and X (mm) is the diameter (distance between load points) of the samples between two load points.

The volatilization rate (H) of the LWA is calculated according to the Cd content measured by the inductively coupled plasma emission spectrometer (ICP-OES) (before the test, the sample should be dried and ground fine, and passed the 200 target sieve). The calculation formula is shown in equation (S1):

$$H = \left(1 - \frac{K(1 - LOI)}{S}\right) \times 100\% \quad \text{Eq.(S1)}$$

In equation (S1), H is the volatilization rate of Cd, K (μg/g) is the Cd content in the sintered sample, S (μg/g) is the Cd content of the samples before sintering, and LOI is the loss on ignition at 1000 ℃, which obtained by X-ray fluorescence spectrometer (XRF).

The leaching test of heavy metal ions was conducted according to the Toxicity Characteristic Leaching Procedure (TCLP). The LWAs were broken into particles of about 5 mm, dried in an oven for 24 hours. Then the extraction solution with a pH at about 2.88 ± 0.05 were prepared and the sample particles were added at a liquid-solid ratio of 20:1 (L/kg). The mixtures were shaken at a rate of 110 ± 10 times/min for 18 hours in a horizontal oscillator, and then left to stand for 2 hours. The supernatant was extracted using a disposable syringe. The impurities were removed through a pinhole filter, and then the ion leaching concentration of heavy metal ions were tested by atomic absorption spectrometer (CONTAA-700).

After the leaching test, the sample was dried at a temperature of 105 ℃ for 24 hours, and the residual heavy metal cadmium content was tested by an inductively coupled plasma emission spectrometer (ICP-OES). The solidified rate (G) of the lightweight aggregate sample is calculated according to equation (S2).

$$G = \frac{C_2}{C_1} \times 100\% \qquad \text{Eq.(S2)}$$

C_1 and C_2 are the Cd content before and after leaching in the LWAs respectively (μg/g).

X-ray fluorescence spectrometer (Zetium-X) was performed to analyze the chemical composition of the raw materials. The XRD was performed by a D8-Advance X-ray diffractometer (XRD, Bruker Optics, Germany) to analyze the mineral phase. The micro-structures of the samples were analyzed by SEM (Quanta 450 FEG) and the elements distribution of the samples in the LWAs were analyzed by SEM-EDS (ESCALAB 250Xi). The valence of different elements in the samples were analyzed by XPS (ESCALAB 250Xi). TG-MS tests are performed in an Ar atmosphere with a heating rate of 10°C/min up to 1300°C to analyze the gas emission from LWAs during high temperature sintering process.

Table S1 Chemical composition of raw materials (wt%)

Raw materials	LOI	SiO_2	Al_2O_3	Fe_2O_3	CaO	K_2O	MgO	Na_2O	SO_3	others
CS	5.31	64.23	15.43	6.72	4.28	1.36	0.82	0.44	0.09	1.32
LS	1.57	34.42	20.76	2.05	9.17	8.16	0.67	8.58	7.97	6.65

Table S2 Different CaO Content of Samples.

CaO content (%)	0.2	0.4	0.6	0.8	1.0
KCl	Cl-LWA-0.2	Cl-LWA-0.4	Cl-LWA-0.6	Cl-LWA-0.8	Cl-LWA-1.0
Na_2SO_4	S-LWA-0.2	S-LWA-0.4	S-LWA-0.6	S-LWA-0.8	S-LWA-1.0

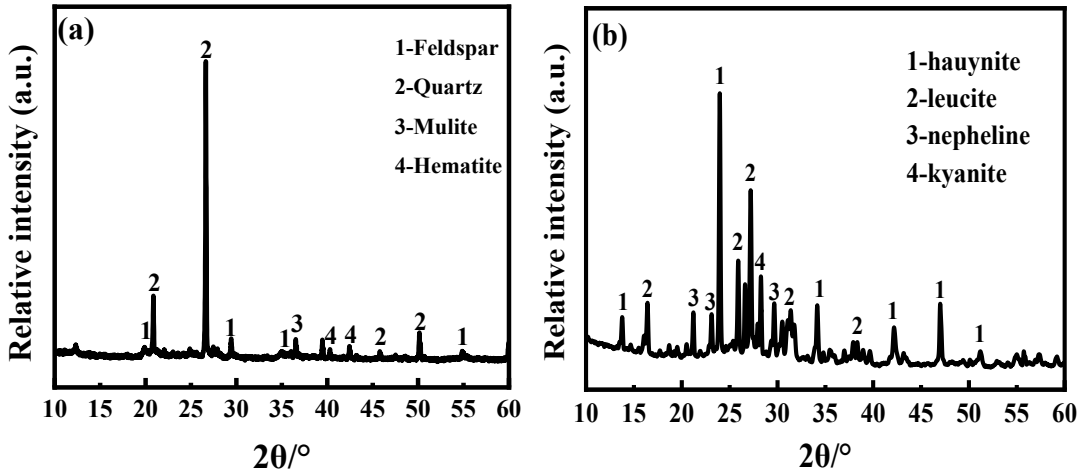

Figure S1 XRD patterns of (a) CS and (b) LS

2. Fitting of Arrhenius dynamics model

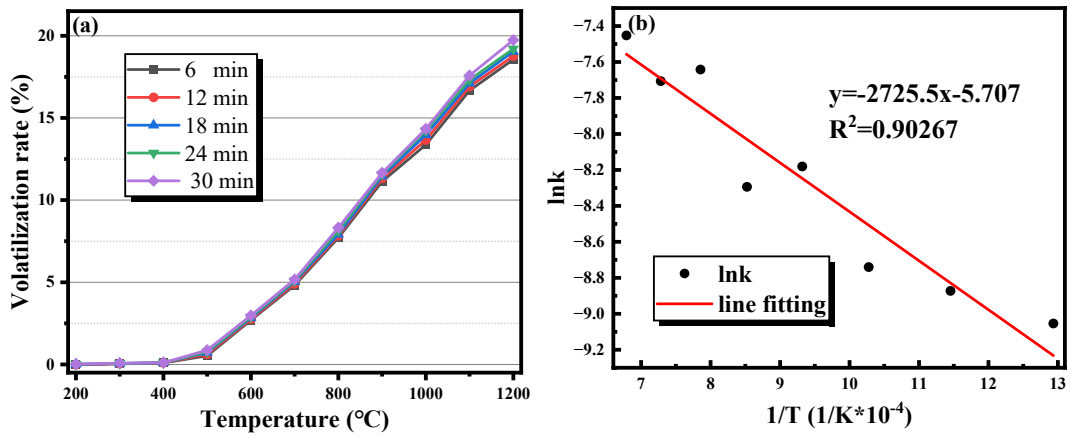

Figure S2. (a) Volatility at different sintering temperatures at different sintering time points and (b) 1/T with lnk fitting results of Cl-LWAs-0

Table S3. Fitting results of -ln(1-H) and t for Cl-LWAs-0

Temperature (℃)	Fitting results	R^2
500	$y = 0.00014x + 0.00447$	0.98493
600	$y = 0.00012x + 0.02653$	0.99506
700	$y = 0.00016x + 0.04852$	0.98447
800	$y = 0.00028x + 0.07819$	0.96434
900	$y = 0.00025x + 0.11696$	0.97281
1000	$y = 0.00048x + 0.14112$	0.96811
1100	$y = 0.00045x + 0.17966$	0.99685
1200	$y = 0.00058x + 0.20109$	0.93858

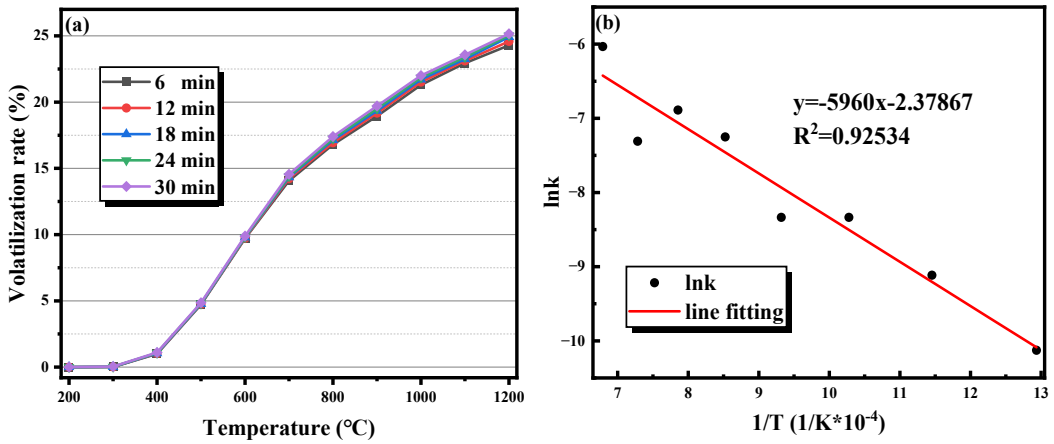

Figure S3. (a) Volatility at different sintering temperatures at different sintering time points and (b) 1/T and lnk fitting results of Cl-LWA-0.8

Table S4. Fitting results of -ln(1-H) and t for Cl-LWAs-0.8

Temperature (℃)	Fitting results	R^2
500	$y = 0.00004x + 0.00454$	0.90415
600	$y = 0.00011x + 0.02019$	0.96007
700	$y = 0.00024x + 0.09842$	0.94098
800	$y = 0.00024x + 0.14799$	0.93876
900	$y = 0.00071x + 0.18091$	0.98374
1000	$y = 0.00102x + 0.00454$	0.97184
1100	$y = 0.00067x + 0.23640$	0.99212
1200	$y = 0.00240x + 0.33065$	0.98994

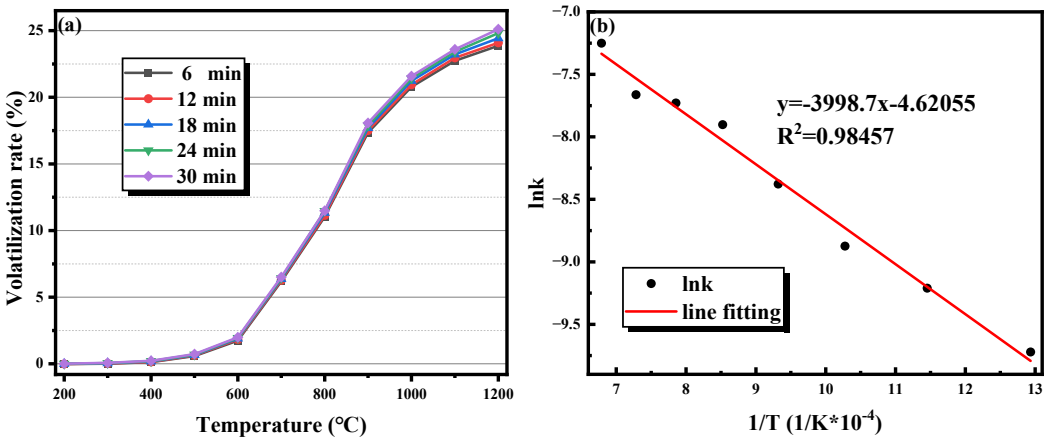

Figure S4. (a) Volatility at different sintering temperatures at different sintering time points and (b) 1/T and lnk fitting results of S-LWAs-0

Table S5. Fitting results of -ln(1-H) and t for S-LWAs-0

Temperature (℃)	Fitting results	R^2
500	$y = 0.00006x + 0.00553$	0.99244
600	$y = 0.00010x + 0.01716$	0.96509
700	$y = 0.00014x + 0.06304$	0.99501
800	$y = 0.00023x + 0.11566$	0.93289
900	$y = 0.00037x + 0.18819$	0.99815
1000	$y = 0.00044x + 0.23028$	0.98957
1100	$y = 0.00047x + 0.25534$	0.99570
1200	$y = 0.00071x + 0.26781$	0.99154

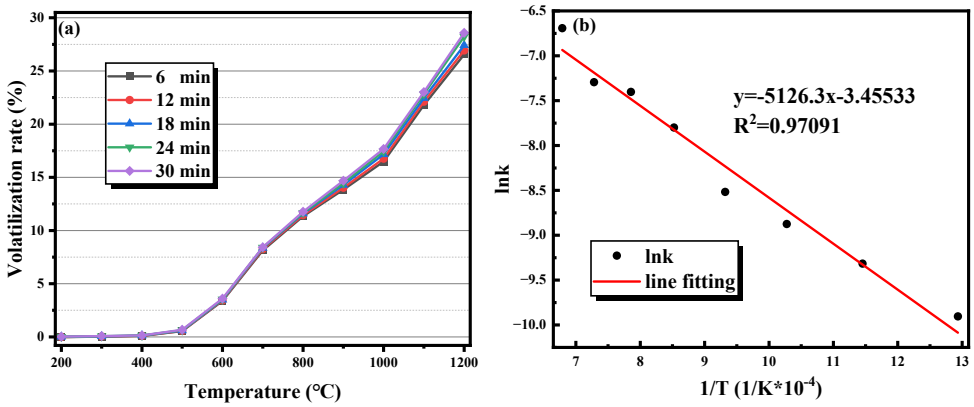

Figure S5. (a) Volatility at different sintering temperatures at different sintering time points and (b) 1/T and lnk fitting results of S-LWAs-0.8

Table S6. Fitting results of -ln(1-H) and t for S-LWAs-0.8

Temperature (℃)	Fitting results	R^2
500	$y = 0.00005x + 0.00514$	0.98631
600	$y = 0.00009x + 0.03383$	0.98807
700	$y = 0.00014x + 0.08429$	0.91131
800	$y = 0.00020x + 0.11927$	0.98417
900	$y = 0.00041x + 0.14645$	0.99550
1000	$y = 0.00061x + 0.17648$	0.98852
1100	$y = 0.00068x + 0.24196$	0.99076
1200	$y = 0.00124x + 0.29996$	0.97609

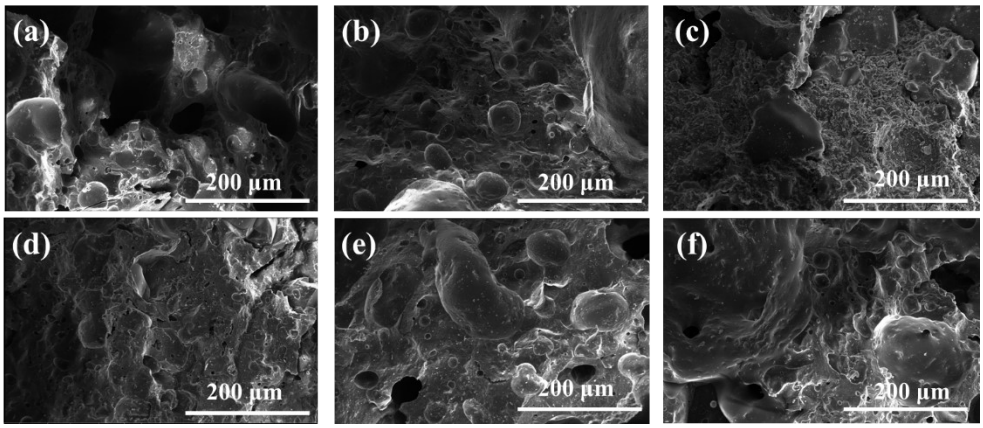

Figure S6. SEM micrographs of the LWA samples.

Table S7. EDS results of elements of Cl-LWA-0 in selected area (wt.%).

Elements	wt%	Wt % Sigma	At%
O	67.23	0.32	78.55
Na	1.20	0.12	0.97
Al	7.17	0.14	4.97
Si	20.67	0.22	13.75
K	3.10	0.11	1.48
Ca	0.56	0.09	0.26
Cd	0.08	0.21	0.01
Total	100.00		100.00

Table S8. EDS results of elements of Cl-LWAs-0.8 in selected area (wt.%).

Elements	wt%	Wt % Sigma	At%
O	55.87	0.37	69.80
Na	1.42	0.11	1.23
Al	9.18	0.16	6.80
Si	26.12	0.26	18.59
K	3.03	0.13	1.55
Ca	3.88	0.14	1.93
Cd	0.50	0.24	0.09
Total	100.00		100.00

Table S9. EDS results of elements of S-LWA-0 in selected area (wt.%).

Elemnts	wt%	Wt % Sigma	At%
O	57.05	0.34	70.76
Na	1.47	0.11	1.27
Al	7.03	0.13	5.17
Si	28.42	0.25	20.08

K	1.64	0.10	0.83
Ca	3.50	0.13	1.73
Cd	0.89	0.23	0.16
Total	100.00		100.00

Table S10. EDS results of elements of S-LWAs-0.8 in selected area (wt.%).

Elements	wt%	Wt % Sigma	At%
O	57.92	0.33	71.53
Na	2.77	0.12	2.38
Al	8.07	0.13	5.91
Si	24.74	0.22	17.40
K	1.98	0.10	1.00
Ca	3.11	0.12	1.54
Cd	1.41	0.21	0.25
Total	100.00		100.00

Table S11. Comparison of the Cd solidification ratio in this work with previously relevant systems.

System	Treatment	Solidification ratio(%)	Leached (mg/L)	References
Municipal solid waste incineration	DFT calculation &adjust pH	98.92	-	[1]
Contaminated soils	Binder preparartion	-	0.620	[2]
Municipal solid waste incineration-fly ash	Vulcanization, precipitation and bio-oxidation	-	0.070	[3]

Lead-zinc tailings	Using highly reactive pozzolanic solid waste	97.05	-	[4]
Municipal solid waste incineration fly ash	MgO-based binders	-	-	[5]
Phosphogypsum slag-based cementitious materials	Carbonization	-	0.00427	[6]
Contaminated soils	Sintering	>99.9	-	This work

References:

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