Supporting Information

Experimental Section

Steel slag sample

The steel slag used in this study was obtained from the steel converter of the Beijing Shougang steel plant in Beijing. The fresh slag, which was sampled after water washing, crushing, and magnetic separation, was ground into smaller particles that could pass through a 200-mesh screen (particle size <74 mm) at ambient temperature in a closed environment, and then kept in the dark in sealed vacuum containers after calcined at 850°C for 10 min in a N_2 atmosphere until use.

Carbonation experiments

Thermogravimetric analysis (TGA) was used to investigate the carbonation–calcination looping of steel slag using a TGA/DSC 1 STARe thermogravimetric analyzer (Mettler-Toledo, Inc., Columbus, OH, USA). Three simulated flue gases according to the typical compositions of an actual flue gas and pure CO_2 were prepared to study the CO_2 capture capacity of the steel slag (Table S1). Firstly, the pretreated steel slag was heated from 200°C to 900°C at a rate of 20 °Cmin^{-1} separately in the four different reaction gases to study the influence of temperature and CO_2 content on the uptake of CO_2 by the steel slag sample. Then the carbonation–calcination looping experiments were conducted according to the following procedure in different reaction gases at atmospheric pressure: carbonation for 5 min at 600°C, then rapid heating at a rate of 100 °Cmin^{-1} to 800°C for a 5–min duration of calcination, and finally cooling down to 600°C at a rate of 50 °Cmin^{-1} for another looping cycle.

During the carbonation experiments, an average weight of 10.5 mg of the pretreated steel slag was placed on an alumina 150 mL pan in a uniform layer of 1 mm and preheated in the N_2 atmosphere with a heating rate of 10 °Cmin^{-1} until the target temperature was achieved; then the atmosphere was instantly switched into a 20 mLmin^{-1} flow of the reaction gas for detection. All carbonation experiments were conducted in duplicate.

Table S1 Compositions of all the gases used in this study

Reaction gas[a]	N_2	O_2	CO_2	H_2O
Simulated flue gas #1	85%	10%	5%	<10 ppm
Simulated flue gas #2	80%	10%	10%	<10 ppm
Simulated flue gas #3	75%	10%	15%	<10 ppm
Gas #4	0	0	100%	<10 ppm

[a] All of the reaction gases in this study were purchased from Beijing ZG Special Gases Science & Technology Co., Ltd. (PR China).

Material characterization

The carbon content of the steel slag was analyzed by using a CE- 440 elemental analyzer (Electronic Associates, Inc., Chelmsford, MA, USA). The composition and content of other elements in the steel slag, which were calibrated against the carbon value for accuracy, were analyzed by X-ray fluorescence (XRF) using an XRF-1800 Analyzer (Shimadzu Corp., Kyoto, Japan). Surface area, particle size, and pore size of the slag sample were determined with an ASAP 2020 HD88 Surface Area and Porosity Analyzer (Micromeritics Co., Norcross, GA, USA).

X-ray diffraction (XRD) analysis of the steel slag was performed on a Smartlab X-ray diffractometer (Rigaku Corp., Tokyo, Japan) with the following operating parameters: CuK_α radiation (λ = 1.5418 Å), FeK_β filter, 40 kV, 200 mA power generator. An angular range of 2θ= 10–60° was measured with a step size of 0.02° and a 2 s counting time per step. The identification of all crystalline phases was undertaken with JADE5 software (Materials Data, Inc., Livermore, CA, USA) and the PDF-2 2004 database (International Centre for Diffraction Data, Newton Square, PA, USA).

Analytical methods

The reference intensity ratio (RIR) method, which is also known as the “matrix flushing” method[1], was used for quantifying the mineral phase containing available Ca for carbonation in the steel slag sample, with corundum (α-Al_2O_3 powder ≥99.95 %; Alfa Aesar, Ward Hill, MA, USA) used as an internal standard. The content of any crystal phase i in the sample, added with a known amount of corundum, is given by Equation (1):

$$x_i = \frac{I_i}{I_{cor}} \cdot \frac{x_{cor}}{k_{i,cor}} \cdot \frac{1}{1 - x_{cor}} \qquad (1)$$

where x is the weight fraction, I is the intensity of the most intense line, and the subscripts i and cor are phase i and the standard phase corundum, respectively. The value $k_{i,cor}$ represents the reference intensity ratio of phase i to corundum, which was determined according to convention from the most intense line of phase i and corundum in a 50:50 mixture by weight.

The carbonation conversion η_{Ca} was determined from the available Ca content of the raw steel slag ($Ca_{available}$ in wt%), the molar weights of Ca (MW_{Ca} in gmol^{-1}) and CO_2 (MW_{CO_2} in gmol^{-1}), and the uptake of CO_2 by dry weight. In Equation (2) the assumption is made that only Ca is carbonated during the carbonation process. And the relative conversion refers to the carbonation conversion of a certain carbonation–calcination looping cycle, divided by the maximum carbonation conversion at the initial cycle.

$$\eta_{Ca}(\%) = \frac{CO_2(wt\ \%) \times \dfrac{MW_{Ca}(g/mol)}{MW_{CO_2}(g/mol)}}{Ca_{available}(g/g)} \times 100 \qquad (2)$$

Elemental and Mineral Composition of the Steel Slag

Table S2 Elemental composition of the steel slag from Beijing Shougang steel plant

Bulk density	Content [%]								
[gcm^{-3}]	H_2O	C	CaO	Al_2O_3	SiO_2	MgO	Fe_2O_3	Mn	Ti
1.29	1.65	0.95	39.68	12.12	10.89	10.05	7.63	0.61	0.38

The main elemental components of the slag sample were CaO, Al_2O_3, SiO_2, and

MgO, which were all above 10% and in total accounted for 72.74 %. In addition, the most abundant heavy metals were Mn and Ti, with Cr, Ba, and Zn also present. The total amount of heavy metals represented approximately 1.2% of the total mass of the slag sample. The slag sample was derived from the steel converter residues after the process of magnetic separation, so the concentration of Fe was not high due to it being sufficiently recycled.

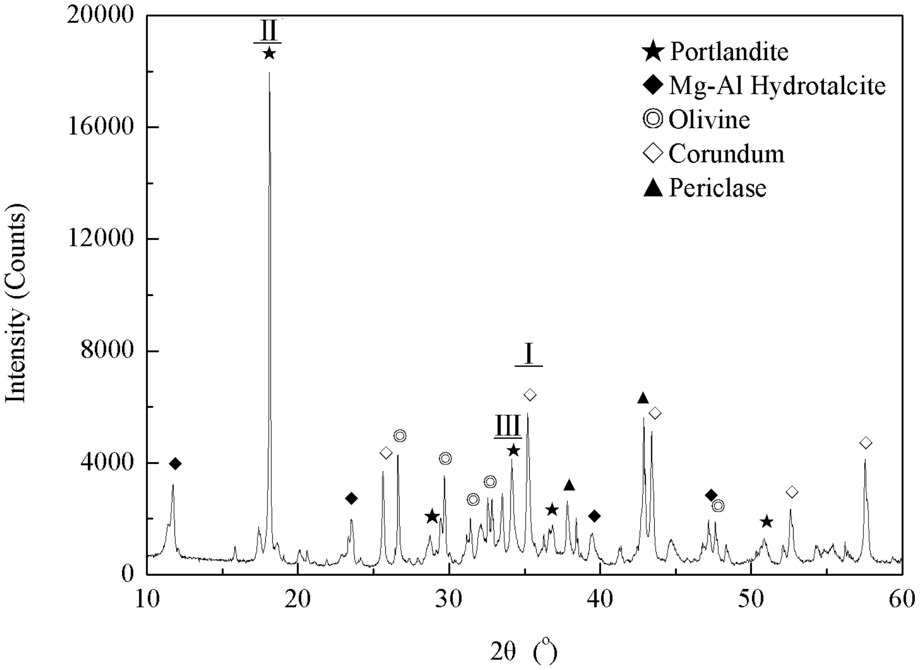

Fig. S1 XRD patterns of the raw steel slag mixed with 20% of corundum.

The XRD pattern of the steel slag sample mixed with 20% corundum is shown in Fig. S1, indicating that the main mineral composition of steel slag was portlandite, Mg–Al hydrotalcite, olivine, and periclase, with other minor phases including andradite, katoite, Ca_2SiO_4, and rutile. The existence of portlandite ($Ca(OH)_2$), which was available for carbonation to sequestrate CO_2, theoretically verified the CO_2 capture potential of steel slag. Using XRD in combination with the RIR method, the steel slag sample was determined to contain 18.95% of portlandite according to Equation (1) (Table S3), where the intensity of the strongest line of corundum (I, 2θ = 35.15°) and the second strongest line of portlandite (II, 2θ = 18.06°) were used. Here, the second strongest line of portlandite (II) was selected to replace its strongest line (III, $2\theta = 34.10°$) in the calculation to avoid a potential overlap between the strongest line of corundum (I) and the strongest line of portlandite (III). One can infer that the steel slag used in this study had a maximum theoretical CO_2 capture potential of 112.7 mg_{CO_2} g_{slag}^{-1}, which was in accordance with the typical values (99–135 mg_{CO_2} g_{slag}^{-1}) for general steel slags.[2]

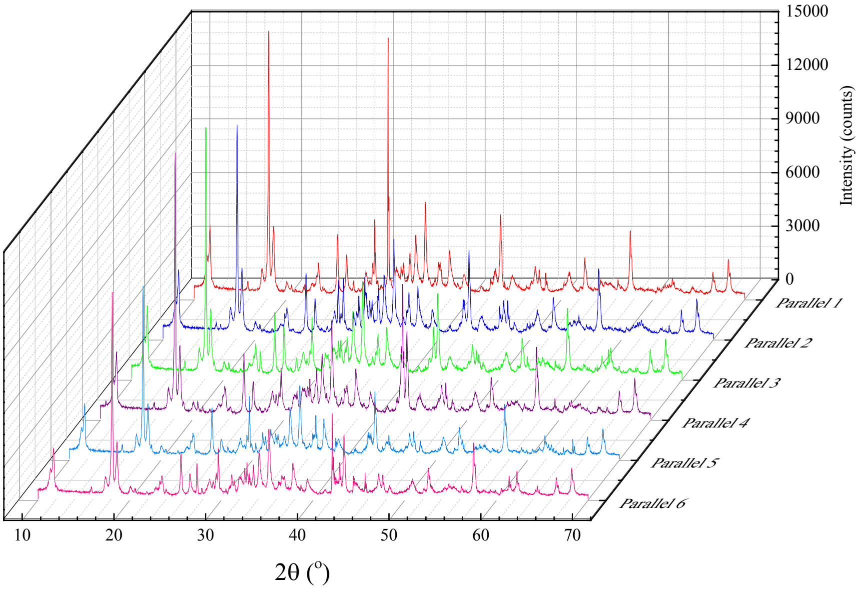

Fig. S2 Parallelism experiments for the calculation of $Ca(OH)_2$ content in the steel slag in Table S3.

Table S3 Quantitative analysis of $Ca(OH)_2$ in the steel slag sample

Repeated exp. no.	$I_{Ca(OH)_2}$:I_{cor}	$K_{Ca(OH)_2,cor}$	Weight fraction (%)	Average weight fraction (%)
1	98871:57141	2.23	19.42	18.95 ± 1.23
2	84121:55676	2.23	16.96	
3	95637:56171	2.23	19.11	
4	100568:57581	2.23	19.60	
5	69359:42876	2.23	18.15	
6	84783:46539	2.23	20.45	

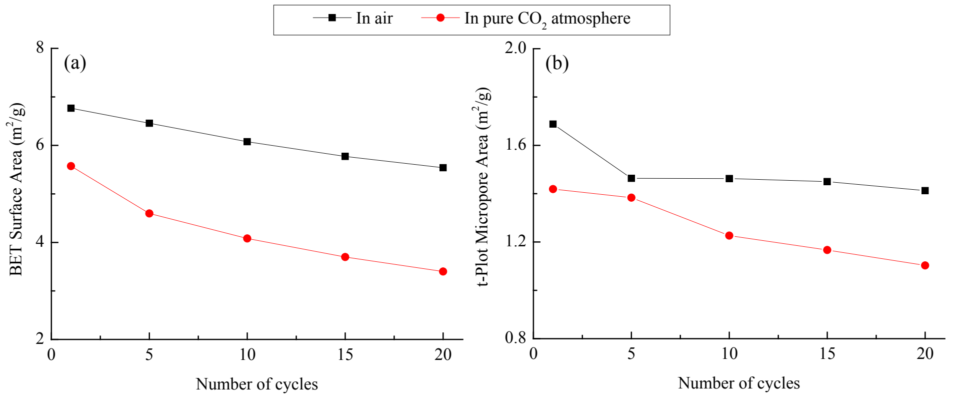

Fig. S3 Change of the surface areas of the steel slag materials with the carbonation–calcination cycle numbers: (a) Nitrogen physical adsorption of Brunauer–Emmett–Teller (BET) surface area at 77K, (b) Nitrogen physical adsorption of Harkins and Jura t-plot micro-pore area at 77K.

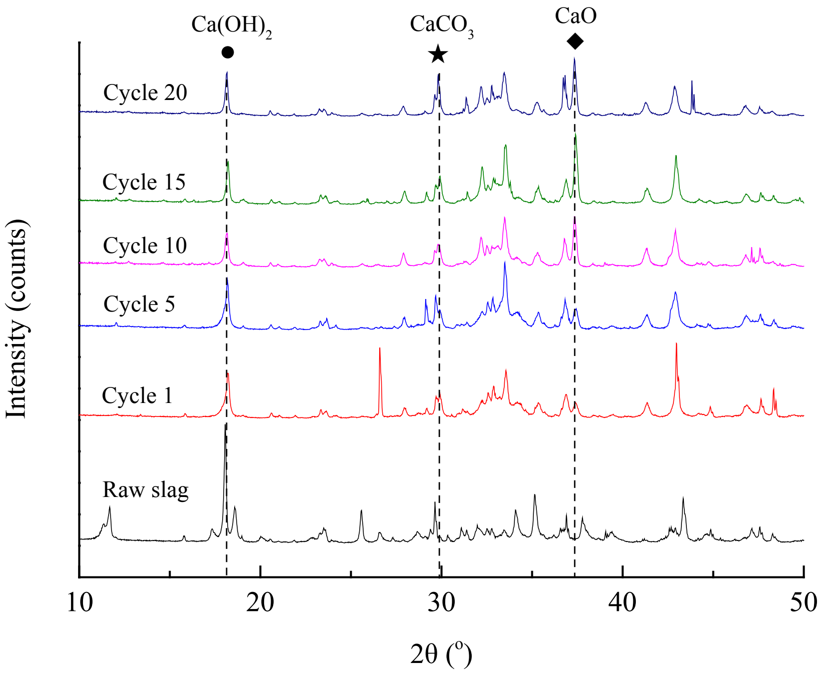

Fig. S4 Range of the calcium-based mineral phases in the steel slag after multiple carbonation–calcination looping cycles.

Notes and references

1 F. H. Chung, *J. Appl. Crystallogr.*, 1974, **7**, 519.

2 P. Renforth, C. L. Washbourne, J. Taylder and D. A. C. Manning, *Environ. Sci. Technol.*, 2011, **45**, 2035.