Supplementary File 1: Supplementary experiment to this study.

Experiment

The analytical grade calcium acetate monohydrate, cerium nitrate hexahydrate, and glacial acetic acid were sourced from Aladdin Reagent (Shanghai) Co., China. The CaO-based materials were prepared by wet-mixing method applied in the previous work[1]. Pre-determined masses of the precursor salts were dissolved in deionized water, agitated at ambient temperature for 0.5 h, and subsequently dehydrated at 120 °C. The dried samples were calcined at 700 °C under air atmosphere in a muffle furnace for 2 h. The resulting samples were labeled as Ni_xCe_y-CaO, where *x* and *y* correspond to the mass percentages of NiO and CeO_2 in the samples. For example, $Ni_{10}Ce_{15}$-CaO contains NiO and CeO_2 at mass fractions of 10% and 15%, respectively. In terms of comparison, the calcium acetate monohydrate was treated at high temperature by a muffle furnace according to the above-mentioned conditions, excluding the addition of CeO_2, and the obtained sorbent was denoted as C-CaO. The cyclic CO_2 capture performances of the CaO-based materials were investigated on a dual fixed-bed reactor. Approximately 300 mg of each material was loaded into a porcelain boat and introduced into the carbonator, where it was exposed to a gas mixture of 15% CO_2/40% H_2O/45% N_2 at 700 °C for 10 min. The carbonated sample was then transferred to the calciner and held at 850 °C under pure N_2 for 10 min. Alternate transportation between the carbonator and calciner was employed t to emulate cyclic CO_2 capture operation. Following each carbonation and calcination step, the porcelain boat was cooled within a vacuum desiccator for 3 min and subsequently weighed on a precision electronic balance. The CO_2 absorption capacity of each material was calculated according to Eq. (6):

$$C_N = \frac{m_{\mathrm{car},N} - m_{\mathrm{cal},N}}{m_0} \tag{6}$$

where, C_N is the CO_2 capture capacity of the material of the *N*th cycle, g/g; $m_{car,N}$ is the mass of the sorbent after the *N*th carbonation, g; $m_{\mathrm{cal},N}$ is the mass of the material after the *N*th calcination, g. The carbonation conversions of the materials were calculated according to Eq. (7):

$$X_N = \frac{m_{\mathrm{car},N} - m_{\mathrm{cal},N}}{m_0} \frac{M_{\mathrm{CaO}}}{M_{\mathrm{CO2}}} \times 100\% \tag{7}$$

where, X_N is the carbonation conversion of the material of the *N*th cycle, %; M_{CaO} and M_{CO2} are the molar masses of CaO and CO_2, respectively, g/mol.

[1] Yan X, Duan C, Yu S, Dai B, Sun C, Chu H. 2024. Revealing the mechanism of oxygen vacancy defect for CO_2 adsorption and diffusion on CaO: DFT and experimental study. *Journal of CO2 Utilization* 79:102648. https://doi.org/10.1016/j.jcou.2023.102648.