

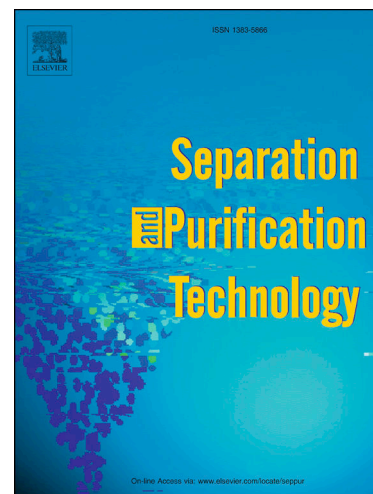
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Efficient-and-Stable CH₄ Reforming with Integrated CO₂ Capture and Utilization using Li₄SiO₄ Sorbent

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Abstract

CO₂ capture and utilization has been considered as an up-and-coming short- to mid-term approach to mitigate the excessive CO₂ emission. Comparing to the conventional separate capture, transportation and conversion arrangement, the integrated CO₂ capture and utilization (ICCU) could largely simplify the complex process and reduce the energy consumption. However, the poor stability of high-temperature CO₂ sorption/desorption severely limit the potential of ICCU. Therefore, it is indispensable to develop a new sorbent/catalyst system ensuring the high-efficiency and long-term operation of the ICCU. In this paper, we propose and demonstrate the feasibility and performance of using K₂CO₃-doped Li₄SiO₄ as an efficient CO₂ sorbent for ICCU operating at a relatively low temperature by dry reforming of methane. Results show that the ratio of H₂/CO produced is stabilized at 1±0.05 in the pre-breakthrough stage, and the duration extends to be 1.6 times of the original value in the cyclic operations, displaying an excellent performance in reaction matching and process stability.

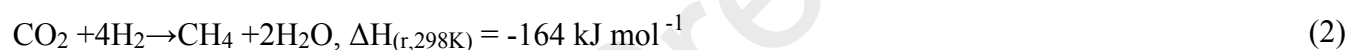
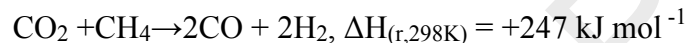
Keywords: Integrated CO₂ capture and utilization; Li₄SiO₄-based sorbent; CO₂ sorption and desorption; Dry reforming of methane

1. Introduction

CO₂ capture and utilization (CCU) has been regarded as a compelling approach to achieve low-carbon and sustainable development of mankind's industry system [1-3]. However, the current CCU process involves separate steps of CO₂ capture and conversion, resulting in high operating cost and energy consumption [4, 5]. Especially for the high-temperature flue gas from power and heat production processes, it needs to be cooled after carbon capture and then heated for CCU process to realize CO₂ utilization, resulting in a serious waste of energy. From the perspective of green and efficient production, integrated operation of the CCU could greatly simplify the process and improve energy efficiency [6, 7].



(1)



(3)

Recently, three reactions as shown in Eq. (1-3) have been reported to achieve the integrated carbon capture and utilization (ICCU) [8-11]. Captured CO₂ can be transformed into CO by reverse water-gas shift (Eq. 1) [12, 13], realizing CO₂ conversion and the produced CO can be further applied in the established Fischer-Tropsch (FT) synthesis [14, 15]. The Sabatier reaction (Eq. 2) uses H₂ and CO₂ as reactants to produce CH₄ and H₂O [16, 17]. Significantly, the methanation reaction is exothermic and can be carried out spontaneously without consuming additional energy, but a large amount of H₂ is consumed. Compared with the above two CO₂ conversion methods, dry reforming of methane (DRM) is more dramatic for converting both greenhouse gases at the same time (Eq. 3) [18, 19]. CO₂ and CH₄ are converted into a mixture of H₂ and CO with a ratio of close to 1, which could be easily converted into liquid chemical

feedstocks via the Fischer-Tropsch (FT) synthesis without additional H_2 [20]. The relatively cheap greenhouse gas reactants and high value-added products make DRM to be one of the most advantageous green chemical approaches for sustainable development.

Some works showed that the desorbed CO_2 could be in-situ converted into CO and H_2 in ICCU by DRM (ICCU-DRM), but the low CO_2 sorption capacity (below $0.05\text{ g }CO_2\text{ g}^{-1}$) is far from meeting the needs of practical application [9, 21]. Recently, lime and $Ni/MgO-Al_2O_3$ were tested in ICCU-DRM as the sorbent and catalyst respectively [6]. It is reported that the ratio of $H_2/CO=1$, which is considered to be suitable for producing long-chain hydrocarbons by FT synthesis, was maintained for 17 min with high conversions of CH_4 and CO_2 (97.1% and 96.3%) in the so-called pre-breakthrough stage. However, in the cyclic stability test, due to the sintering of CaO, only a 50% performance of ICCU-DRM was left after 5 cycles. Generally, a common problem of CaO-based sorbents is the rapid loss of capacity due to sintering [22, 23]. Therefore, it is necessary to explore new CO_2 sorbents with good capacity while satisfying high-temperature stable cyclic sorption and desorption for ICCU-DRM. Similar to CaO, Li_4SiO_4 -based sorbents are also commonly used for CO_2 sorption and desorption at high temperatures [24, 25]. Although the sorption capacity of Li_4SiO_4 -based sorbents ($0.367\text{ g }CO_2\text{ g}^{-1}$) is not as high as that of CaO-based sorbents ($0.786\text{ g }CO_2\text{ g}^{-1}$), Li_4SiO_4 -based sorbents could still play a significant part due to the excellent stability. However, pure Li_4SiO_4 has poor sorption performance at low CO_2 concentration. Modification of Li_4SiO_4 is needed to improve its CO_2 sorption performance, including the treatment by organic acids [26], alkali improvement [27] and doping of other metals [28, 29]. In particular, K_2CO_3 -doped Li_4SiO_4 shows excellent sorption performance and cycling stability at low CO_2 concentrations, suggesting its potential as a sorbent for ICCU-DRM with long-term stability.

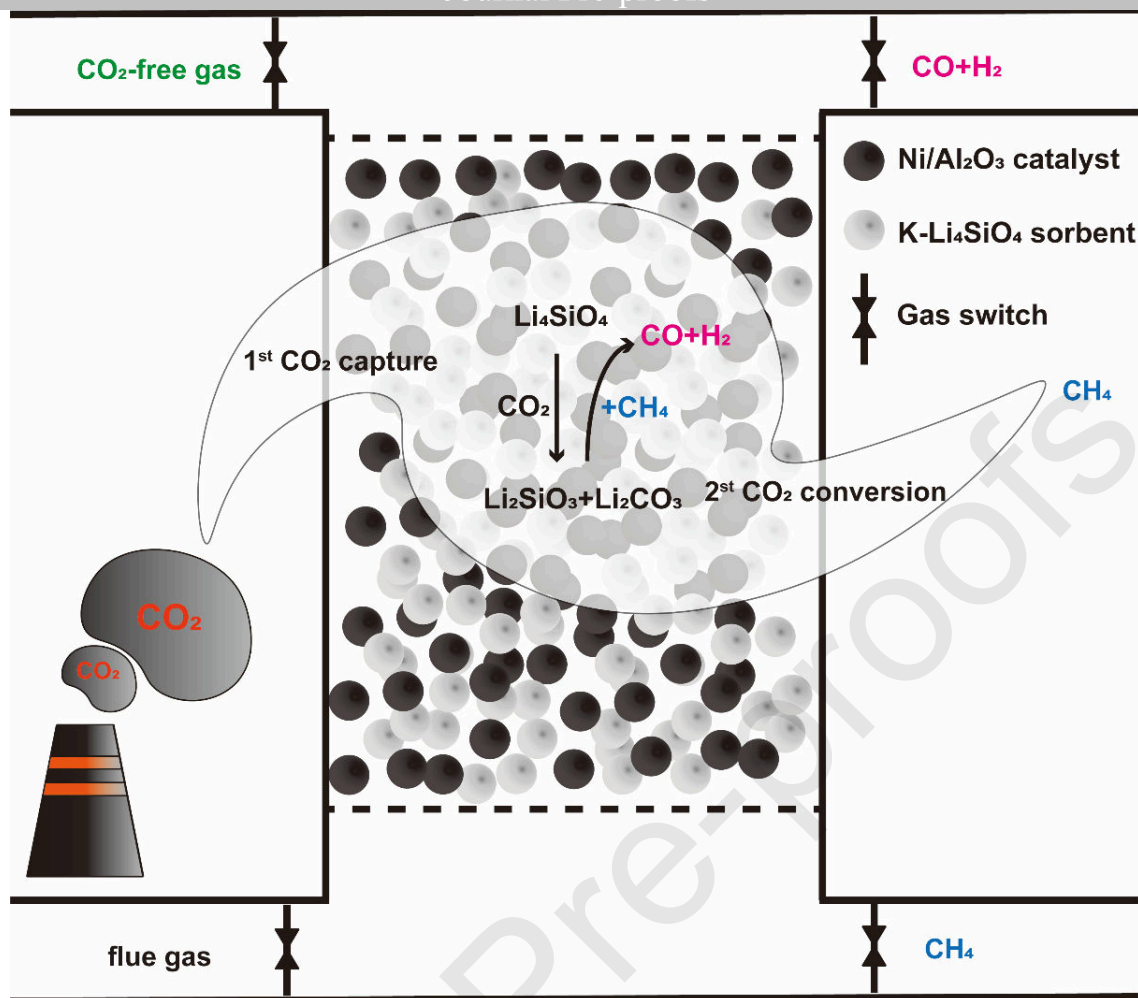


Figure 1. Elementary diagram of the ICCU-DRM process for carbon capture and convert captured CO₂ into CO and H₂ with CH₄.

Herein, for the first time, we propose and demonstrate the feasibility and performance of using K₂CO₃-doped Li₄SiO₄ [30] as the sorbent and Ni/Al₂O₃ as the catalyst to carry out ICCU-DRM in a single reactor isothermally. The steps of the ICCU-DRM process are depicted in Figure 1, including the carbon capture from the flue gas using K₂CO₃-doped Li₄SiO₄ and the conversion of captured CO₂ into H₂ and CO through DRM. In the ICCU-DRM process. The sorption capacity of K₂CO₃-doped Li₄SiO₄ was continuously increased in the first five cycles, making the duration of stable H₂/CO ratio (1±0.05) at pre-breakthrough

stage increase to 1.6 times of the initial value and then stabilize in the following cycles, displaying an excellent stability and great application potential.

2. Experimental

2.1. Sorbent/catalyst preparation

The K-Li₄SiO₄ sorbent was synthesized via a typical solid phase method using Li₂CO₃ ($\geq 99.0\%$ purity, Aladdin), fumed silica (lyophobic, Aladdin) and K₂CO₃ (AR, Aladdin) as precursors. Firstly, SiO₂, Li₂CO₃ and K₂CO₃ was blended with the mole ratio of 1:2.05:0.1 and then fully blended with roll ball for 1 h by a planetary ball grinder (XQM-2) (alternate positive and negative turns, 10 min each time with a speed of 450 r min⁻¹). The materials were collected and calcined by heating at 750 °C for 6 h, and subsequently grinding and sieving to 30-40 mesh.

In a typical experiment, activated alumina (Al₂O₃, 40-60 mesh GC, Aladdin) and Ni(NO₃)₂·6H₂O were used as precursors to prepare 10 wt.% Ni-catalyst by wet impregnation method. After impregnation of Ni(NO₃)₂·6H₂O on activated alumina, the sample was calcined by heating at 550°C for 5 h after drying overnight at 100°C. The obtained catalyst was wetting and sieved to 40-60 mesh.

2.2. Materials characterization

Scanning electron microscope (Quattro S) was used for characterizing the morphology of the sorbent and catalyst. The porous texture and surface area of the sorbents and catalysts were measured by an automatic N₂ sorption analyzer (Quadratorb 2MP). The chemical composition and crystalline phase of samples were analyzed by powder X-ray diffraction (PANalytical X'Pert Powder), and the diffraction pattern was recorded in the 2 θ range of 5-90°. CO₂ sorption/desorption properties of K-Li₄SiO₄ was tested by temperature ramping at a rate of 5 °C min⁻¹ under 15 vol.% CO₂ and cyclic CO₂ sorption capacity of

K-Li₄SiO₄ in 30 cycles was tested using a thermogravimetric analyzer (TGA, NETZSCH STA 2500) at 625 °C as well.

Temperature-programmed reduction in hydrogen (H₂-TPR) and temperature programmed desorption in hydrogen (H₂-TPD) were carried out by a chemisorption apparatus (VDSorb-9Xi). For H₂-TPD, 50 mg catalyst was reduction at 650 °C for 1 h in 10 vol.% H₂/Ar (10 mL min⁻¹). Then it was cooled to 300 °C and kept for 2 h in pure argon (10 mL min⁻¹), and followed by H₂ sorption at 50 °C for 5 h. Finally, H₂ desorption was conducted from 50 °C to 900 °C at a rate of 10 °C min⁻¹. For H₂-TPR, after dehydrated the 50 mg calcined catalyst at 300 °C in Ar (10 mL min⁻¹) and maintained for 1 h, the sample was cooled to 50 °C and the atmosphere was varied into 10 vol.% H₂/Ar (10 mL min⁻¹). Waiting for 30 min at 50 °C, the temperature was increased to 900 °C at a rate of 5 °C min⁻¹.

2.3. Integrated CO₂ capture and utilization test

The tests of ICCU-DRM were performed in a fixed bed with a quart pipe reactor (inner diameter of 20 mm and 400 mm in length) heating by a furnace and the schema of the experimental test system is presented in Fig. S1. Accurate mass flow controllers were used to adjust the feeding of CH₄, CO₂, and N₂. Meanwhile, the concentrations of CH₄, CO₂, CO and H₂ in the exhaust gas were detected continuously by an online gas analyzer (NOVA-975PA) equipped with non-dispersive infrared detectors and thermal conductivity detector. In a typical experiment, the blended sample containing 2 g K-Li₄SiO₄ sorbent and 2 g catalyst were added in quart pipe reactor and preliminarily the reduction of 2 g catalyst was performed at 650 °C for 1 h under 10 vol.% H₂/N₂ (500 ml min⁻¹). Subsequently, the atmosphere was switched into 15 vol.% CO₂/N₂ for simulate flue gas (500 ml min⁻¹) and CO₂ capture was performed at 600 °C, 625 °C or 650 °C for 0.5 h. After CO₂ capture, the gas was changed into N₂ (500 ml min⁻¹) for 2 min so as to purge the reactor under the same temperature of carbon capture, and then the gas was varied into mixture

of CH₄ and N₂ with specified volume concentration (1.7 vol.% CH₄ for 600 °C, 2.1 vol.% CH₄ for 625 °C, and 2.4 vol.% CH₄ for 650 °C) under the same flow velocity for CO₂ desorption and conversion.

The conversion rates (CO₂ and CH₄) and yields (CO and H₂) in the pre-breakthrough stage were calculated as follows:

$$n_{\text{CO}_2}^{\text{desorbed}} = n_{\text{CO}_2}^{\text{unconversion}} + n_{\text{CO}}^{\text{out}} / 2 \quad (4)$$

$$\text{CO}_2 \text{ conversion}(\%) = \int_0^t (n_{\text{CO}_2}^{\text{desorbed}} - n_{\text{CO}_2}^{\text{unconversion}}) / n_{\text{CO}_2}^{\text{desorbed}} \quad (5)$$

$$\text{CH}_4 \text{ conversion}(\%) = \int_0^t (n_{\text{CH}_4}^{\text{feed}} - n_{\text{CH}_4}^{\text{unconversion}}) / n_{\text{CH}_4}^{\text{feed}} \quad (6)$$

where “0” represents the start time of the pre-breakthrough stage and “t” is the end time of the stage for CO₂ conversion.

$$\text{H}_2 \text{ yields}(\%) = \frac{n_{\text{H}_2}^{\text{out}}}{n_{\text{CH}_4}^{\text{feed}} \times 2} \times 100 \quad (7)$$

$$\text{CO yields}(\%) = \frac{n_{\text{CO}}^{\text{out}}}{n_{\text{CH}_4}^{\text{feed}} + n_{\text{CO}_2}^{\text{desorbed}}} \times 100 \quad (8)$$

3. Results and Discussion

3.1. Characteristics of K-Li₄SiO₄ sorbent and Ni/Al₂O₃ catalyst

Morphology and structure of the Ni/Al₂O₃ and K-Li₄SiO₄ are presented in Fig. 2. The K-Li₄SiO₄ sorbent is observed to have a coral-like appearance, and a large number of light spots in element mapping indicates that K is successfully doped in Li₄SiO₄. XRD resulted in Fig. 2(c) show that Li₄SiO₄ is the main phase in the sorbent and there is also a small amount of Li₂SiO₃ and Li₂CO₃. Correspondingly, it is seen to have an amorphous bulk morphology of Ni/Al₂O₃ catalyst and the successful adhesion of Ni to alumina, and their structures are also confirmed by XRD in Fig. 2(c). Fig. 2(d) shows that Ni/Al₂O₃ catalyst has a very large specific surface area (144.48 m² g⁻¹) and pore volume (0.441 cm³ g⁻¹), which is conducive to the

dispersion of nickel metal and the contact of gases during reactions. In contrast, the specific surface area and pore volume of K-Li₄SiO₄ are very smaller, only 1.21 m² g⁻¹ and 0.006 cm³ g⁻¹. The pore size distribution curves in Fig. 2(e) depict that Ni/Al₂O₃ catalyst presents a single peak at around 5-20 nm, while the pore size of K-Li₄SiO₄ concentrates at 3-10 nm.

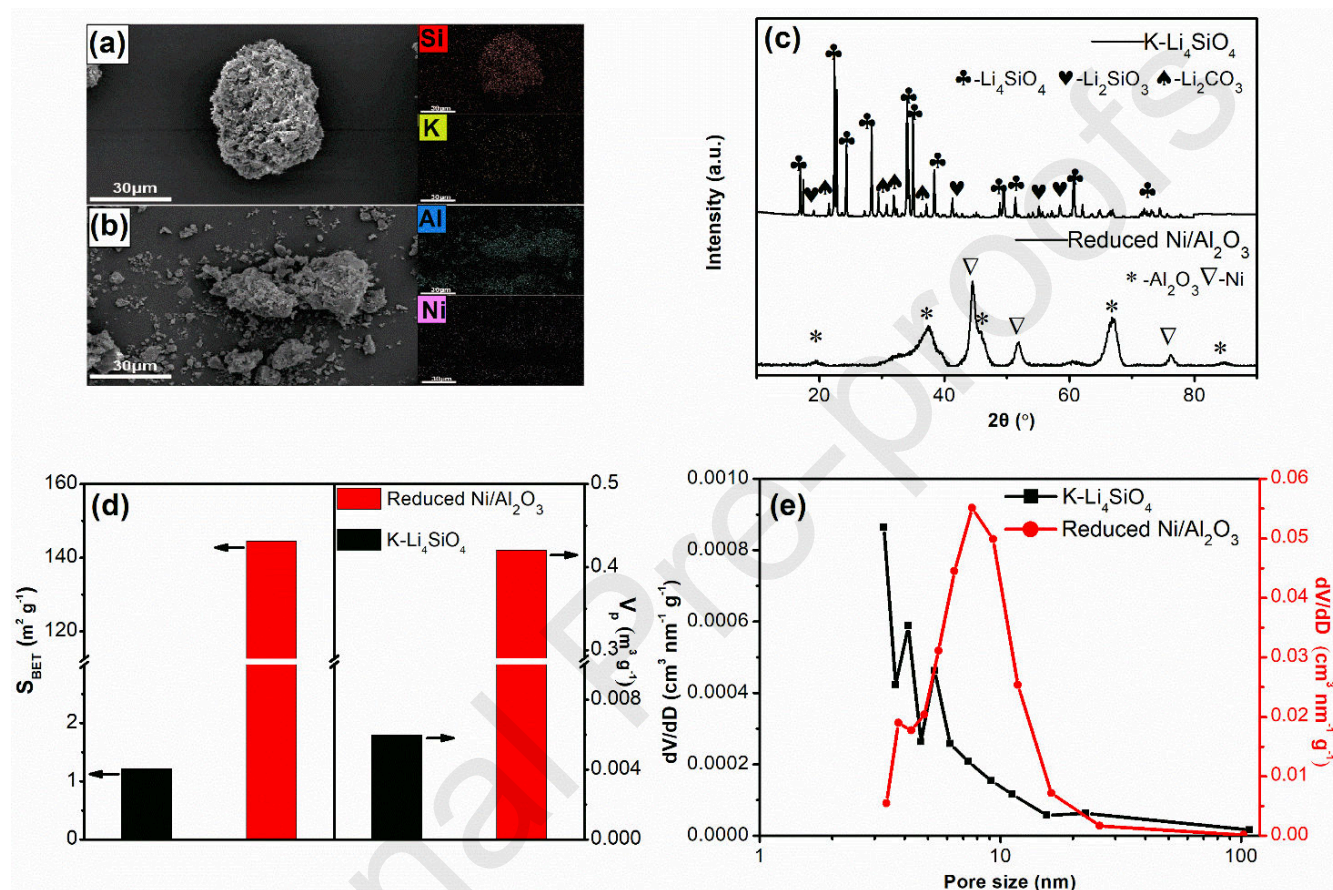


Figure 2. Materials characterization: (a) SEM-EDS images of K-Li₄SiO₄; (b) SEM-EDS images of reduced Ni/Al₂O₃; (c) XRD of K-Li₄SiO₄ and reduced Ni/Al₂O₃; (d) surface area and pore volume of K-Li₄SiO₄ and reduced Ni/Al₂O₃; (e) pore distribution of K-Li₄SiO₄ and reduced Ni/Al₂O₃.

H₂-TPR and H₂-TPD curves of Ni/Al₂O₃ catalyst are shown in Fig. 3(a). The H₂-TPR curve demonstrates three obvious peaks centered at 180 °C, 650 °C and 800 °C, respectively. Combined with the H₂-TPD profile, the peak at 180 °C is owing to the desorption of H₂, which is come from the sorption

of H_2 during temperature rising. The main peak at about 650 °C present the reduction of moderately dispersed small NiO particles. The peak at 800 °C indicates a strong interaction between the NiO specie and activated alumina, resulting in a poorly active phase of $NiAl_2O_4$ [31]. The excellent long-term working stability of the catalyst is reflected in the dry reforming reaction of Ni/Al_2O_3 under 2.1 vol% CO_2 , 2.1 vol% CH_4 for 10h, shown in Fig. S2. The initial CO_2 and CH_4 conversion rates were 86% and 95%, respectively, and the conversion rate dropped by only 1% after 10 h. In addition, the ratio of H_2/CO has been maintained at 1.05-1.07 during 10 h.

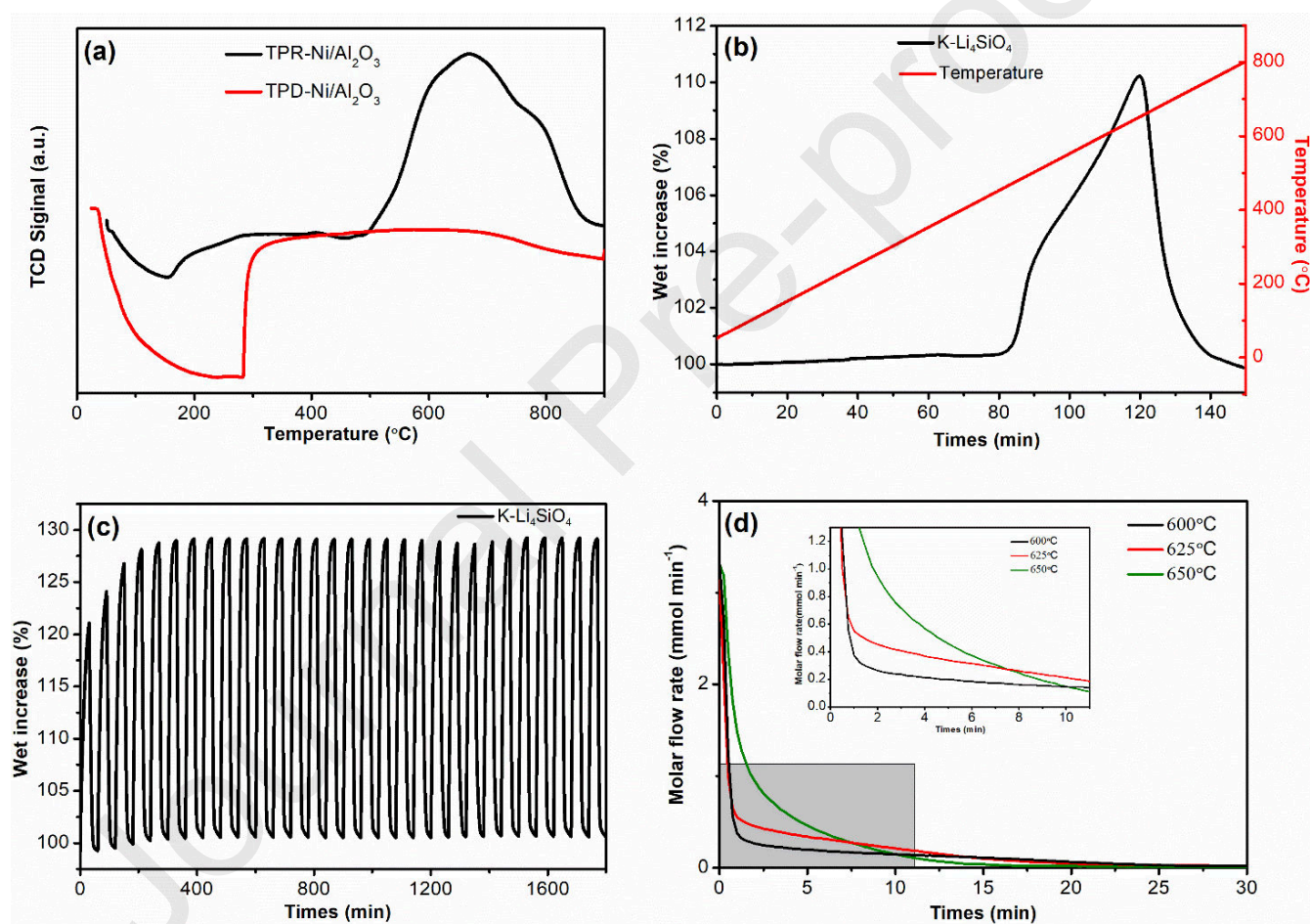


Figure 3. (a) H_2 -TPD and H_2 -TPR of Ni/Al_2O_3 ; (b) Dynamic CO_2 sorption/desorption properties of $K-Li_4SiO_4$ sorbent under 15 vol.% CO_2 ; (c) cyclic CO_2 sorption capacity of $K-Li_4SiO_4$ for 30 cycles at 625 °C on TGA; (d) desorption of $K-Li_4SiO_4$ sorbent at different temperatures in a fixed bed.

For purpose of understand the sorption and desorption properties of $\text{K-Li}_4\text{SiO}_4$, dynamic test was conducted and the curves are displayed in Fig. 3(b). It is quite evident that the sorption of CO_2 starts at $500\text{ }^\circ\text{C}$ and peaks at about $630\text{ }^\circ\text{C}$. The cyclic sorption/desorption curve depicts an increase of CO_2 capacity from the original $0.20\text{ g CO}_2\text{ g}^{-1}$ to $0.27\text{ g CO}_2\text{ g}^{-1}$ after 5 cycles, which is remained in the following 25 cycles. It means that the sorbent possesses an excellent cyclic sorption/desorption stability. Based on these results, three temperatures ($600\text{ }^\circ\text{C}$, $625\text{ }^\circ\text{C}$ and $650\text{ }^\circ\text{C}$) were decided to test the desorption properties of $\text{K-Li}_4\text{SiO}_4$ in the fixed bed to simulate the actual application effect, and the result is exhibited in Fig. 3(d). It is known that the desorption velocities under three temperatures all have fast-then-slow profiles. However, at $650\text{ }^\circ\text{C}$, the CO_2 desorption velocity decreases very rapid, which could not provide stable CO_2 output for the following dry reforming. In contrast, the desorption velocity of CO_2 decreases more slowly at $625\text{ }^\circ\text{C}$ and $600\text{ }^\circ\text{C}$. Nevertheless, stable CO_2 output is only one of the indices for the realization of ICCU-DRM, and the reasonable matching of CO_2 desorption and conversion needs to be further explored.

3.2. Integrated CO_2 capture and utilization property

The ICCU-DRM property of the blended $\text{K-Li}_4\text{SiO}_4$ and $\text{Ni/Al}_2\text{O}_3$ under difference temperatures is exhibited in Fig. 4, which involved three steps: the CO_2 capture, the purge and the CO_2 conversion. Accompanied by the release of CO_2 by $\text{K-Li}_4\text{SiO}_4$ sorbent, the CO_2 conversion stage can be subdivided into the pre-breakthrough and breakthrough stage. For pre-breakthrough stage, since the ratio of desorbed CO_2 to CH_4 in the mixed gas is close to 1, the obtainment of H_2 and CO through dry reforming is stable. Later, the CO_2 conversion enters the breakthrough stage on account of the reduction of the CO_2 desorption velocity. As CO_2 desorption velocity is seriously affected by temperature, the duration of pre-breakthrough stage is different under various temperatures. At $650\text{ }^\circ\text{C}$ as shown in Fig. 4(c), there is

almost no pre-breakthrough stage in the in-situ conversion due to the rapid decrease of CO₂ desorption velocity. At the beginning of the conversion, the amount of CO₂ is much higher than CH₄, causing the occurrence of a significant side reaction ($\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$) and making the amount of produced H₂ is less than that of CO. Gradually, the content of CO₂ becomes lower due to the dramatic reduction of CO₂ release velocity, resulting in methane decomposition ($\text{CH}_4 \rightarrow \text{C} + 2\text{H}_2$) and a H₂/CO ratio bigger than 1. When reaction temperature is set at 625 °C, as presented in Fig. 4(a), the release velocity of CO₂ decreases gently, and the pre-breakthrough time is about 7 min in the in-situ conversion process. Within this stage, the ratio of H₂/CO in product is stable in the range of 0.95-1.05 and the conversion of CH₄ and CO₂ is 95% and 81%, respectively. It should be noted that the continuous consumption of CO₂ by in-situ DRM accelerates the desorption of CO₂ from K-Li₄SiO₄ following Le Chatelier's principle, and results in a much more release of CO₂ in the pre-breakthrough stage during ICCU-DRM than that without the presence of catalyst. Therefore, the actual ratio of CO₂/CH₄ is higher than the pre-set value, which reduces the CO₂ conversion rate significantly. Further decreased reaction temperature into 600 °C, the release of CO₂ becomes more gradual, and the time of pre-breakthrough stage extends to 15 min. However, as the temperature is too low to efficient conversion of CH₄, the conversion of CH₄ and CO₂ during the pre-breakthrough stage is only 75% and 85%, respectively. In conclusion, the optimal operating temperature of K-Li₄SiO₄ and Ni/Al₂O₃ is 625 °C in ICCU-DRM. When the temperature is at 650 °C, CO₂ release velocity of K-Li₄SiO₄ drops too fast to be integrated with the reforming. On the other side, Ni/Al₂O₃ catalyst cannot efficiently convert CO₂ and CH₄ at 600 °C.

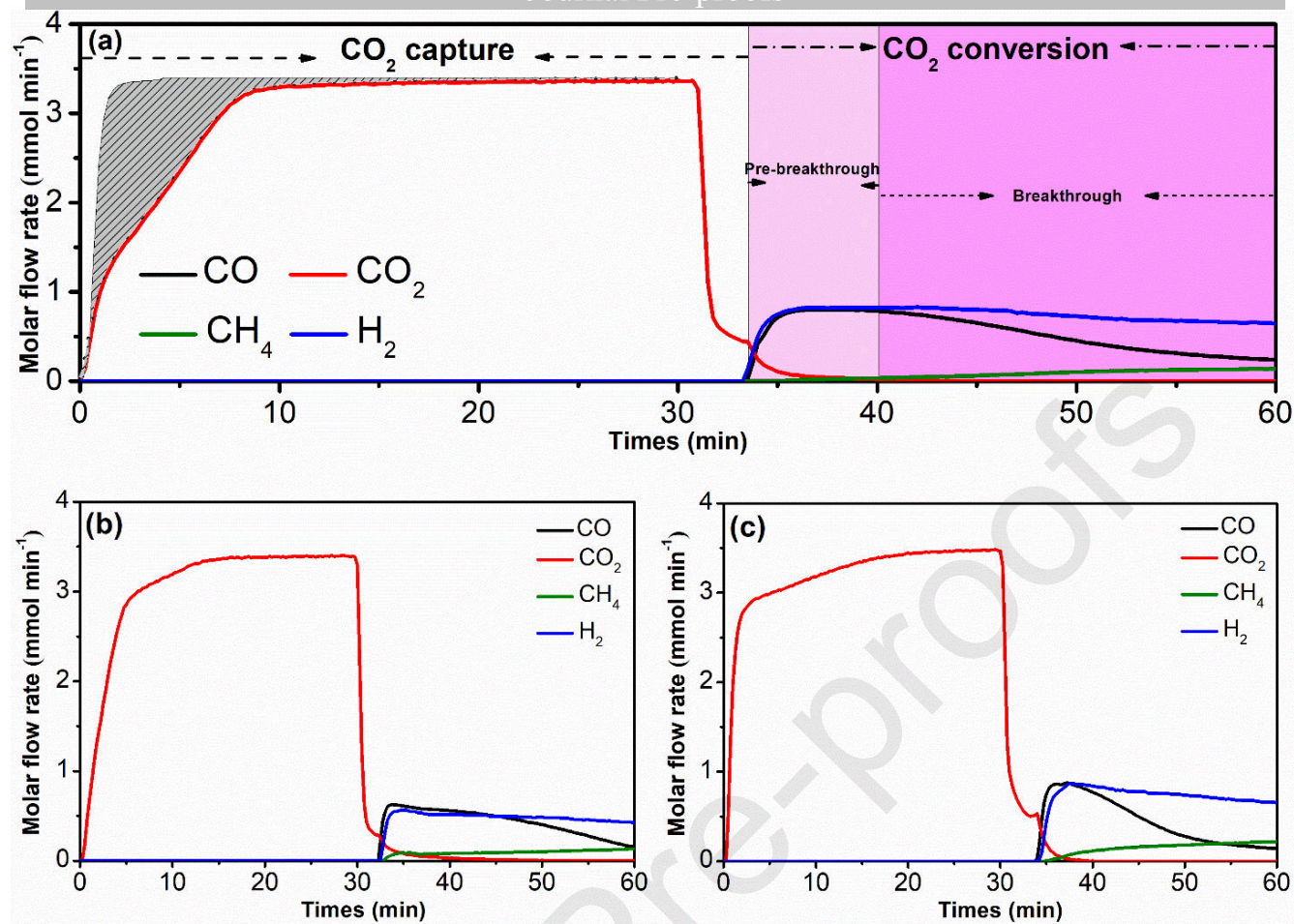


Figure 4. ICCU-DRM at (a) 625 °C; (b) 600 °C; (c) 650 °C.

3.3. Cyclic performance of integrated CO₂ capture and utilization

Based on the optimal coupling temperature of 625 °C, the cyclic performance of ICCU-DRM using K-Li₄SiO₄ and Ni/Al₂O₃ was further tested and characterized. The reaction curves at the 1st, 5th, and 10th cycle are exhibited in Fig. 5(a). In the CO₂ capture stage of the 5th, and 10th cycle, the generation of CO gas is observed on account of the removal of carbon deposits ($\text{CO}_2 + \text{C} \rightarrow 2\text{CO}$) on the catalyst, which mainly comes from methane decomposition of the previous cycle [6]. The ratio of H₂/CO and the molar flow velocities of H₂ and CO as a function of cycle number are further displayed in Fig. 5(b-d). It is noticed that the duration of pre-breakthrough stage is increased from 7 min in the 1st cycle to 11 min in the 10th cycle from Fig. 5(b), indicating an excellent matching and stability of the material system of

Ni/Al₂O₃ and K-Li₄SiO₄. Fig. 5(c) demonstrates three nearly overlapping curves of H₂ molar flow velocity in relation to the 1st, 5th and 10th cycle of ICCU-DRM indicating that the catalyst possesses an excellent cyclic stability during ICCU-DRM process. Furthermore, Fig. 5(d) depicts that the molar flow velocity of CO in the 5-20 min stage rises as growing the number of cycles, which should be credited to the gradual activation of the sorbent in the first five cycles. Meanwhile, the conversion of CO₂ during breakthrough stage increases from 81% to 84%, though there is a slight reduction in the conversion of CH₄ from 95% to 94%. Additionally, though there is a little lower CO₂ conversion than the reported 96.3% at 720 °C by lime and Ni/MgO-Al₂O₃ in the work of Kim et al. [6], a much superior stability on the CO₂ capacity, pre-breakthrough duration and H₂/CO ratio in the cyclic reactions is demonstrated in the work. More information on the comparison of our work with the results in the literature can be found in Table S2.

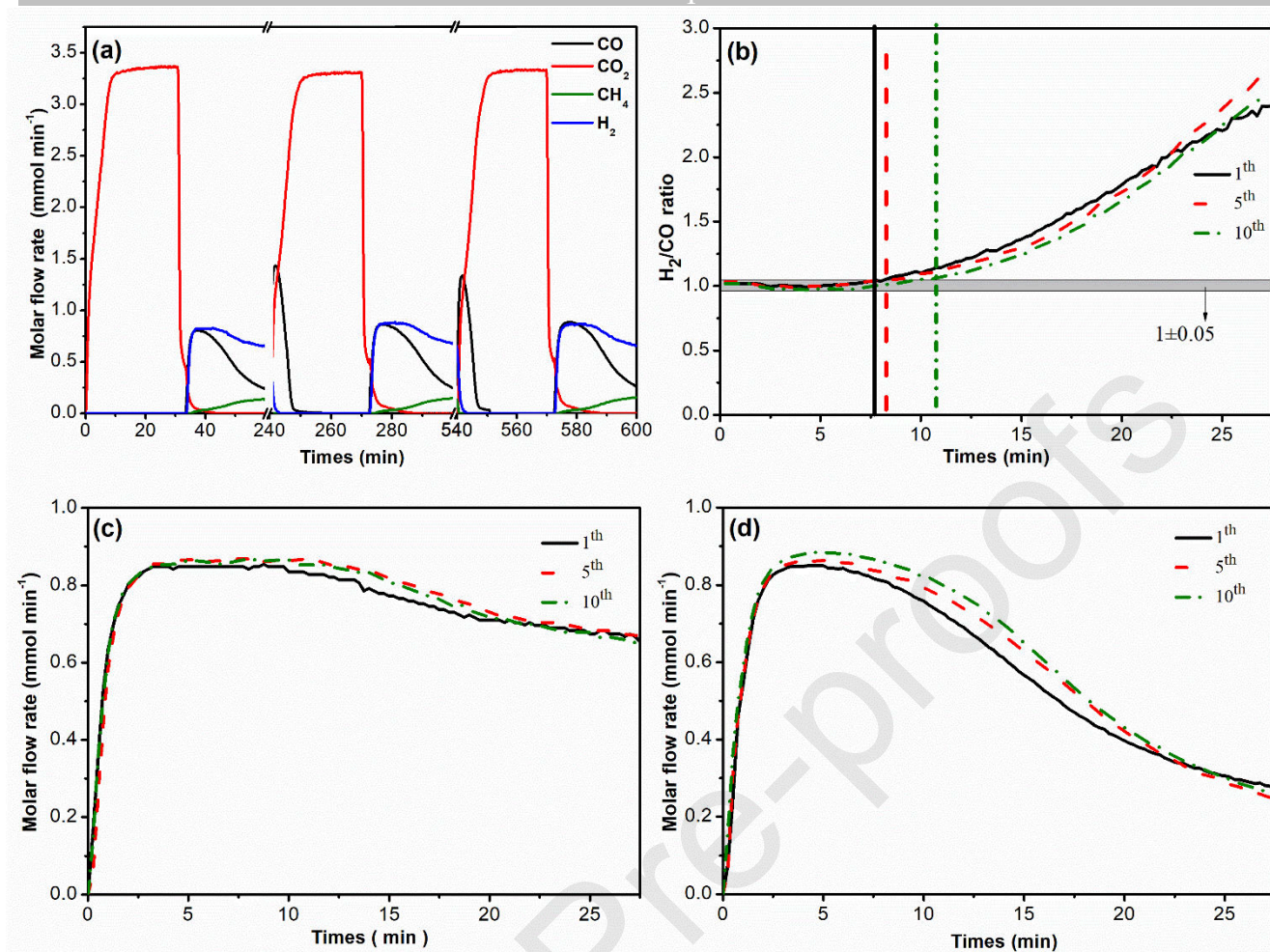


Figure 5. (a) ICCU-DRM at the 1st, 5th and 10th cycle; (b) H₂/CO ratio of the effluent gas; (c) molar flow rate of H₂; (d) molar flow velocity of CO.

SEM images of K-Li₄SiO₄ and Ni/Al₂O₃ after 10 cycles are revealed in Fig. 6(a, b). It is found that the morphology of K-Li₄SiO₄ and Ni/Al₂O₃ remains unchanged after 10 ICCU-DRM cycles. However, some Ni is detected distributing on the surface of K-Li₄SiO₄, indicating the emergence of Ni transfer from Ni/Al₂O₃ to K-Li₄SiO₄ during the ICCU-DRM process [32]. Phase compositions of the sorbent and catalyst after 10 cycles was also analyzed by XRD and presented in Fig. 6(c). It can be found that Li₄SiO₄ is still the main phase in K-Li₄SiO₄, but there is some LiAlO₂ appearing in the Ni catalyst, indicating very slight reaction of Li from the sorbent with Al₂O₃ in the catalyst during the high temperature contact in ICCU-

DRM process. The micro-structural parameters of the fresh $\text{Ni}/\text{Al}_2\text{O}_3$ and $\text{K-Li}_4\text{SiO}_4$ and those after 10 cycles are summarized in Fig. 6(c-d) and Table S1 as well. While a reduction of the pores ranging in 4-20 nm is observed of the $\text{Ni}/\text{Al}_2\text{O}_3$ catalyst after 10 cycles, the pore size distribution of $\text{K-Li}_4\text{SiO}_4$ is almost the same. Generally, the morphology and phase composition of $\text{K-Li}_4\text{SiO}_4$ sorbent and $\text{Ni}/\text{Al}_2\text{O}_3$ catalyst remain basically unchanged after 10 cycles, showing good cyclic stability. But Li and Ni transfer between $\text{K-Li}_4\text{SiO}_4$ sorbent and $\text{Ni}/\text{Al}_2\text{O}_3$ catalyst may be another factor affecting their working life.

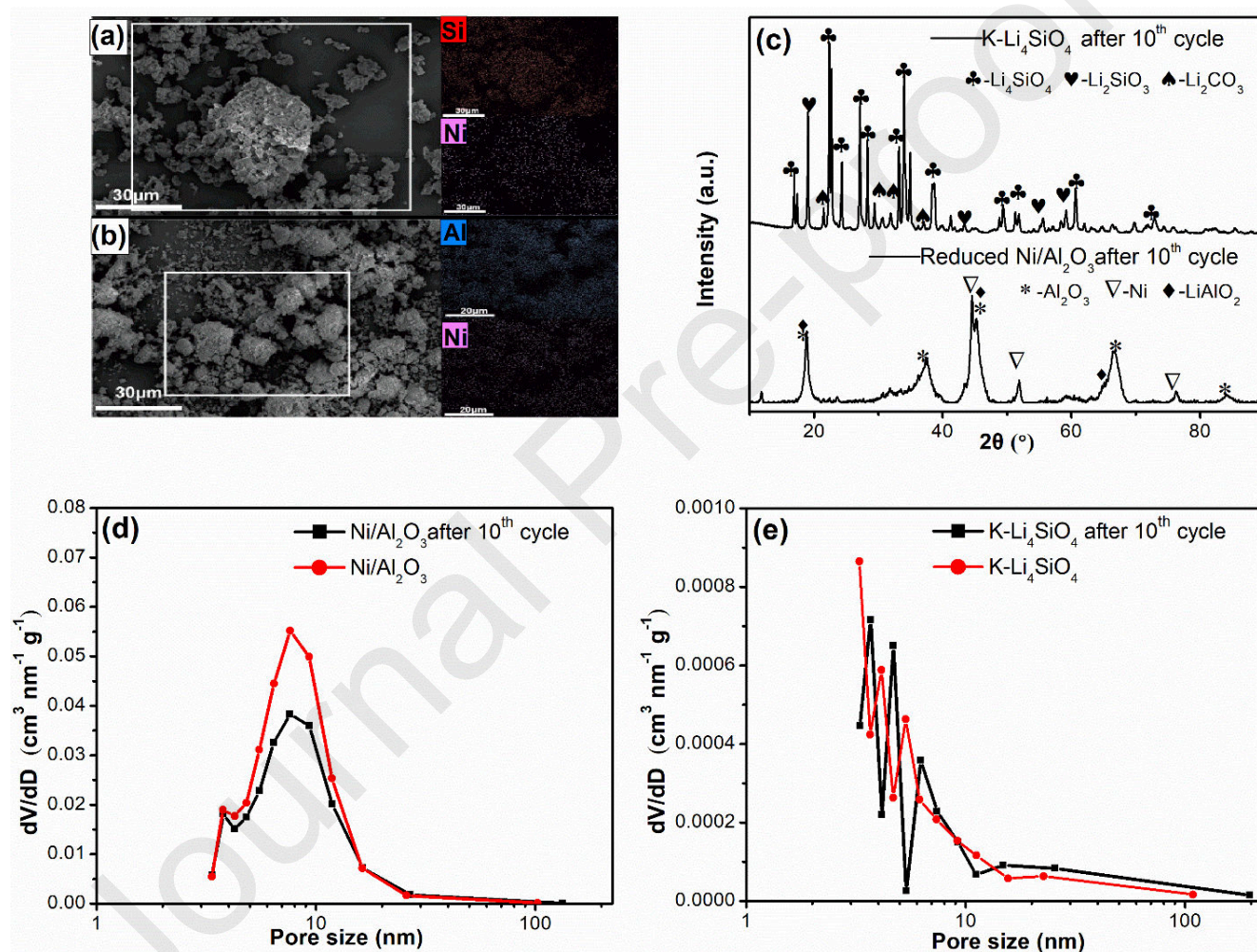


Figure 6. Materials characterization: (a) SEM-EDS images of $\text{K-Li}_4\text{SiO}_4$ after 10 cycles; (b) SEM-EDS images of reduced $\text{Ni}/\text{Al}_2\text{O}_3$ after 10 cycles; (c) XRD patterns of $\text{K-Li}_4\text{SiO}_4$ and reduced $\text{Ni}/\text{Al}_2\text{O}_3$ after 10 cycles; (d) pore distribution of fresh $\text{Ni}/\text{Al}_2\text{O}_3$ and $\text{Ni}/\text{Al}_2\text{O}_3$ after 10 cycles; (e) pore distribution of fresh $\text{K-Li}_4\text{SiO}_4$ and $\text{K-Li}_4\text{SiO}_4$ after 10 cycles.

4. Conclusions

In summary, a good material system containing K-Li₄SiO₄ sorbent and Ni/Al₂O₃ catalyst was successfully applied in the ICCU-DRM process. By the integration of DRM with CO₂ capture, the captured CO₂ is directly converted to syngas with an approximate equimolar ratio of H₂ to CO in a single reactor at a relatively low temperature (625 °C). Though very slight interaction between the highly active Li in K-Li₄SiO₄ with Al₂O₃ in Ni/Al₂O₃ is observed, the material system still shows very excellent cyclic stability during 10 cycles tested, and the duration of stable H₂/CO ratio at about 1±0.05 gradually extends as growing the number of ICCU-DRM cycles. To strengthen the great potential of the coupled K-Li₄SiO₄ and Ni/Al₂O₃ in practical industrial applications of ICCU-DRM, fabrication of the bifunctional composite material containing Li₄SiO₄ and Ni will be the focus in the next step.

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Supporting Information

Schematic of the experimental test system; Physical properties of the CO₂ sorbent and catalyst before and after use.

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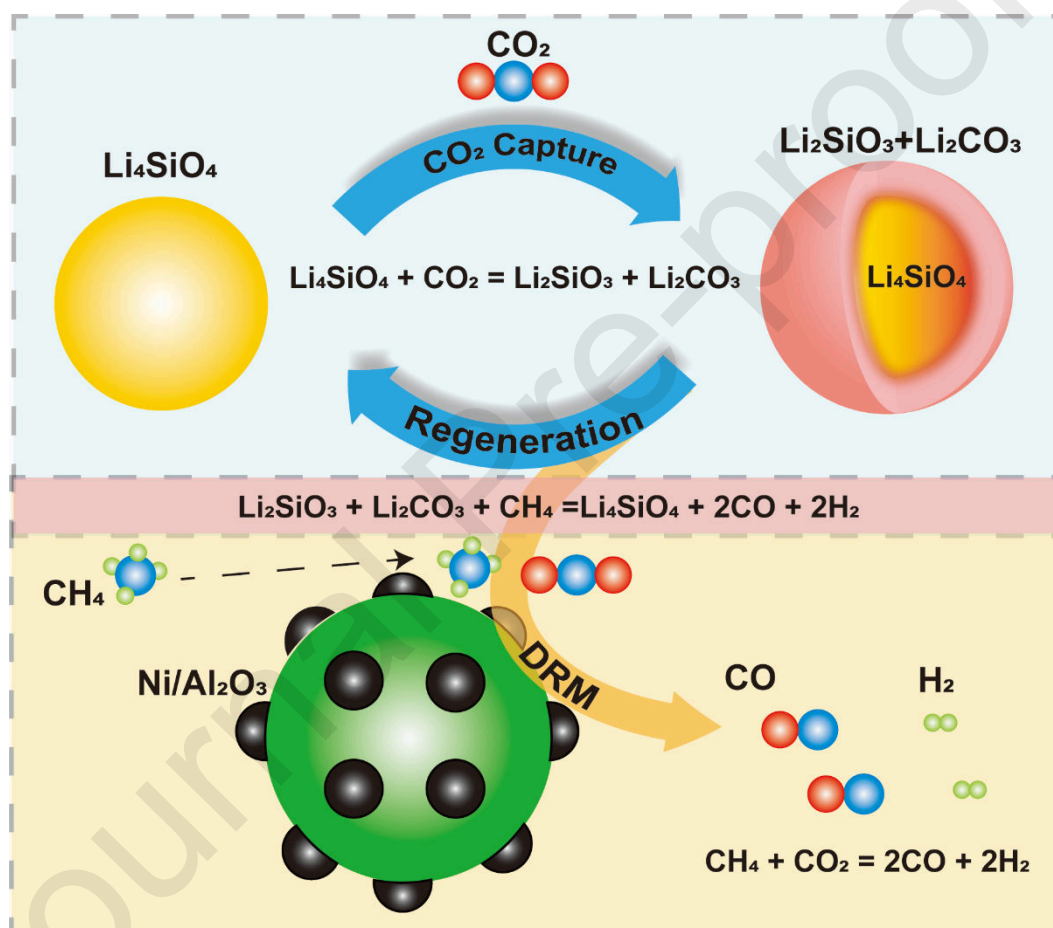
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Highlights

- Proposal of a new material system for the integrated CO₂ capture and utilization.
- Process matching of the CO₂ desorption and in-situ dry reforming of CH₄.
- Demonstration of excellent cyclic stability of ICCU-DRM using Li₄SiO₄ sorbent.

Graphical Abstract



Declaration of Interest Statement:

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled.

Credit author statement:

Zongze Lv: Conceptualization, Formal analysis, Investigation, Validation, Writing - Original Draft.

Changlei Qin: Conceptualization, Resources, Formal analysis, Project administration, Supervision, Writing - Review & Editing, Funding acquisition. **Shuzhen Chen:** Formal analysis, Investigation. **Dawid**

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Efficient-and-Stable CH₄ Reforming with Integrated CO₂ Capture and Utilization using Li₄SiO₄ Sorbent

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