Supplementary Information (SI) for Sustainable Energy & Fuels. This journal is © The Royal Society of Chemistry 2025

## **Supporting information**

Multi-stage structured catalyst system for post-treatment of GHG exhausted from industrial processes

Choji Fukuhara<sup>a</sup>\* Hiroto Naiki<sup>a</sup>, Hiroshi Akama<sup>a</sup>, Yuki Yamada<sup>a</sup>, Priyanka Verma<sup>b</sup>, Ryo Watanabe<sup>\*</sup>

- a) Department of Applied Chemistry and Biochemical Engineering, Graduate School of Engineering, Shizuoka University, 3-5-1 Johoku, Chuo-ku, Hamamatsu, Shizuoka, 432-8561 Japan
- b) Department of Chemical Engineering, Faculty of Engineering, Kyushu University, 744 Motooka Nishi-ku, Fukuoka-shi, Fukuoka 819-0395, Japan

Figure S1 Fast Fourier Transform (FFT) patterns of as-prepared  $\rm Ru/CeO_2$  catalyst for  $\rm CO_2$  methanation.

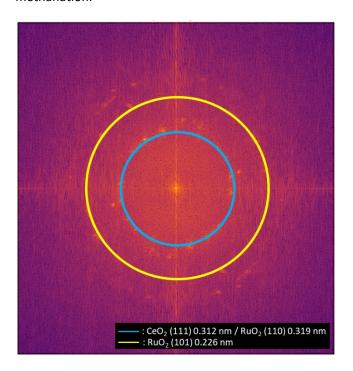


Figure S2 Fast Fourier Transform (FFT) patterns of as-prepared Ni/CeO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> catalyst for DRM.

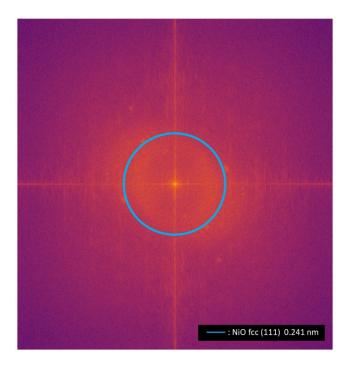
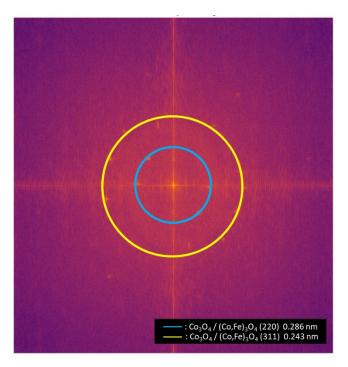


Figure S3 Fast Fourier Transform (FFT) patterns of as-prepared Fe-Co-K catalyst for carbon capture.



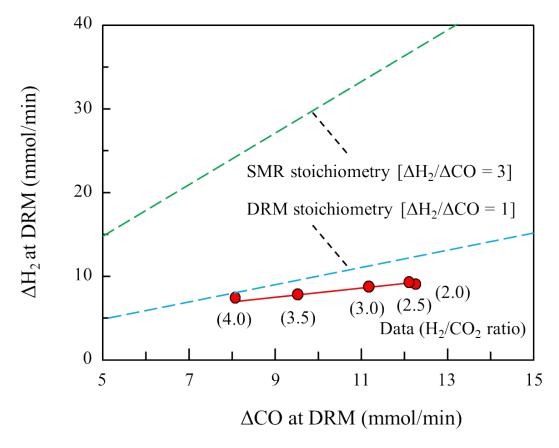


Figure S4 Incremental molar changes of  $H_2$  ( $\Delta H_2$ ) plotted against those of CO ( $\Delta$ CO) in the DRM reactor. Dashed lines represent the stoichiometric reference slopes for DRM ( $\Delta H_2/\Delta$ CO = 1, blue) and SMR ( $\Delta H_2/\Delta$ CO = 3, green).

Table S1 Weight change before and after boring.

	Α	В	С	D	total
Amount of carbon before boring / g	2.23	1.08	0.24	0.15	3.70
Amount of carbon after boring / g	0.93	0.53	0.13	0.08	1.67
Weight change / g	-1.30	-0.55	-0.11	-0.07	-2.03

Table S2 Weight change before and after boring.

	Α	В	С	D	total
Amount of carbon before boring / g	1.72	1.88	1.03	0.44	5.07
Amount of carbon after boring / g	1.41	1.36	0.53	0.11	3.41
Weight change / g	-0.31	-0.52	-0.50	-0.33	-1.66

Table S3 Amount of captured carbon removed through boring operation.

	Α	В	С	D
First boring operation / g	1.30	0.55	0.11	0.07
Second boring operation / g	0.31	0.52	0.50	0.33
Total / g	1.61	1.07	0.61	0.40

Table S4 Amount of catalyst metal removed through boring operation.

Carbon capture position	Removed catalyst / mg	
	48	
$\square$ $\square$ $\square$ $\square$ $\square$ $\square$	32	
	18	
	12	

Figure S5 Reaction characteristic of carbon capturing system by using reuse catalyst.

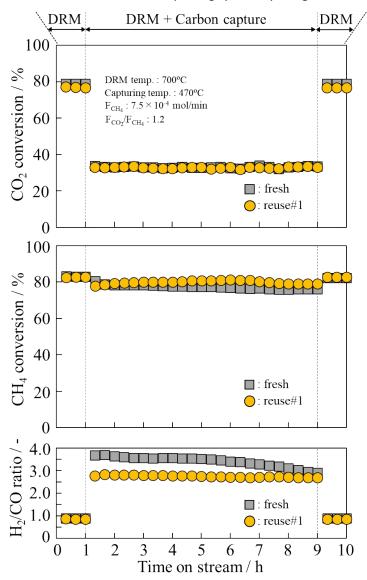


Figure S6 Amount of carbon over reuse catalyst.

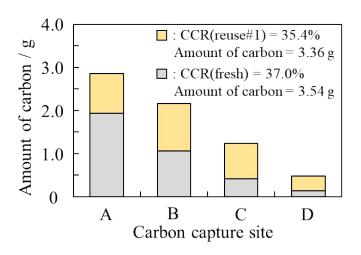


Figure S7 Captured carbon over reuse catalyst.

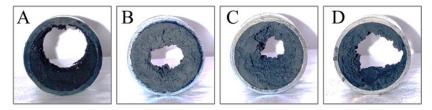


Figure S8 Reaction characteristic of carbon capturing system by using reuse#2 catalyst.

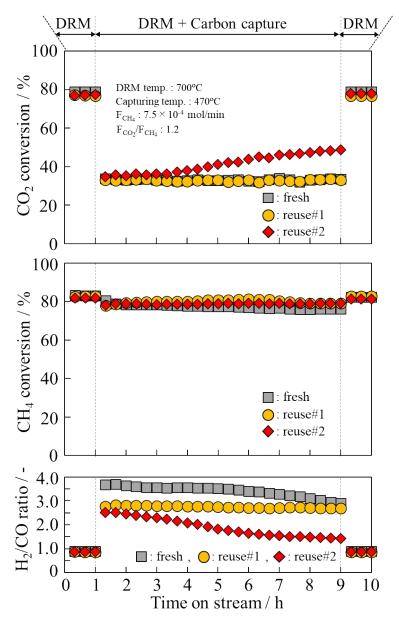


Figure S9 Amount of carbon over reuse#2 catalyst.

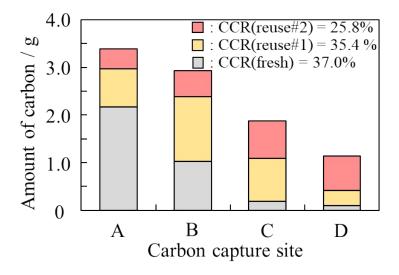


Figure S10 Captured carbon over reuse#2 catalyst.

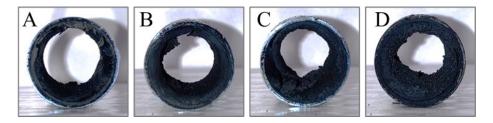


Figure S11 Weight loss curves measured by TG-DTA over captured carbon.

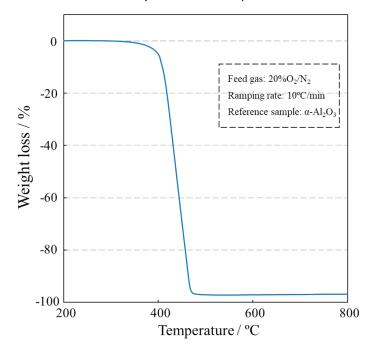


Figure S12 Reaction characteristic of carbon capturing system by using reuse#3 catalyst.

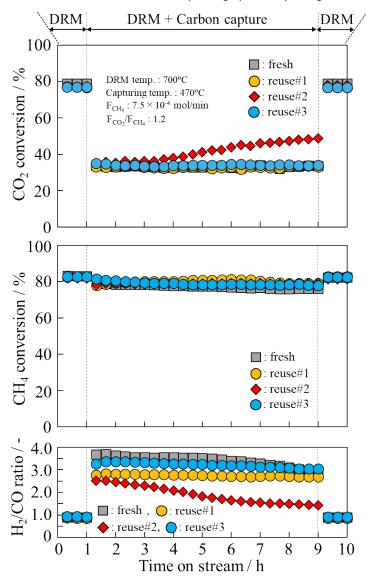


Figure S13 Amount of carbon over reuse#3 catalyst.

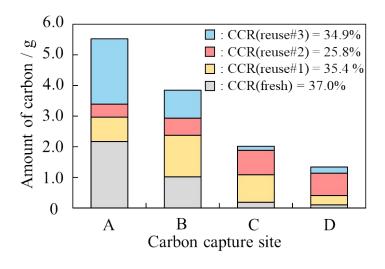


Figure S14 Captured carbon over reuse#3 catalyst.

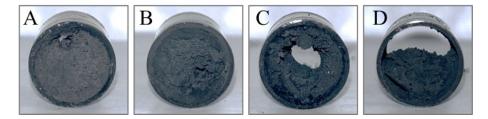


Figure S15 Overall exergy balance of the integrated methanation–DRM–carbon capture system considering external energy inputs. Exergy values at the outlet of each reaction stage are shown for  $H_2/CO_2$  feed ratios of 2.0, 2.5, 3.0, 3.5, and 4.0.

