Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Unveiling Anomalous CO_2-to-N_2 Selectivity of Graphene Oxide

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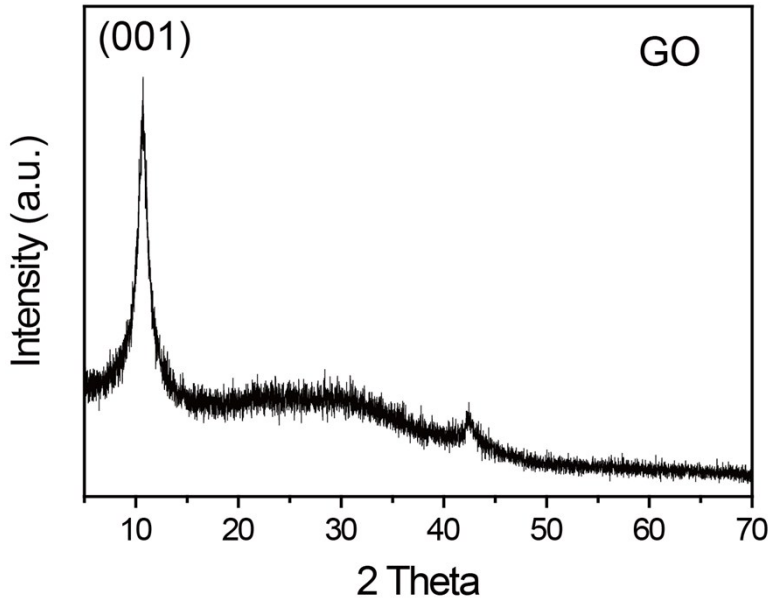

Fig. S1 X-ray diffraction pattern of GO.

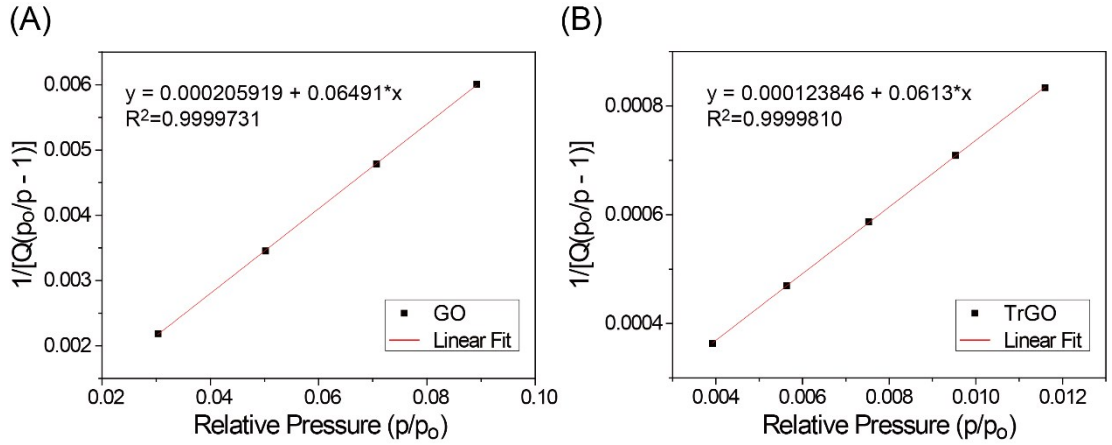

Fig. S2 BET linear plots for (A) GO and (B) TrGO from their corresponding Ar isotherms.

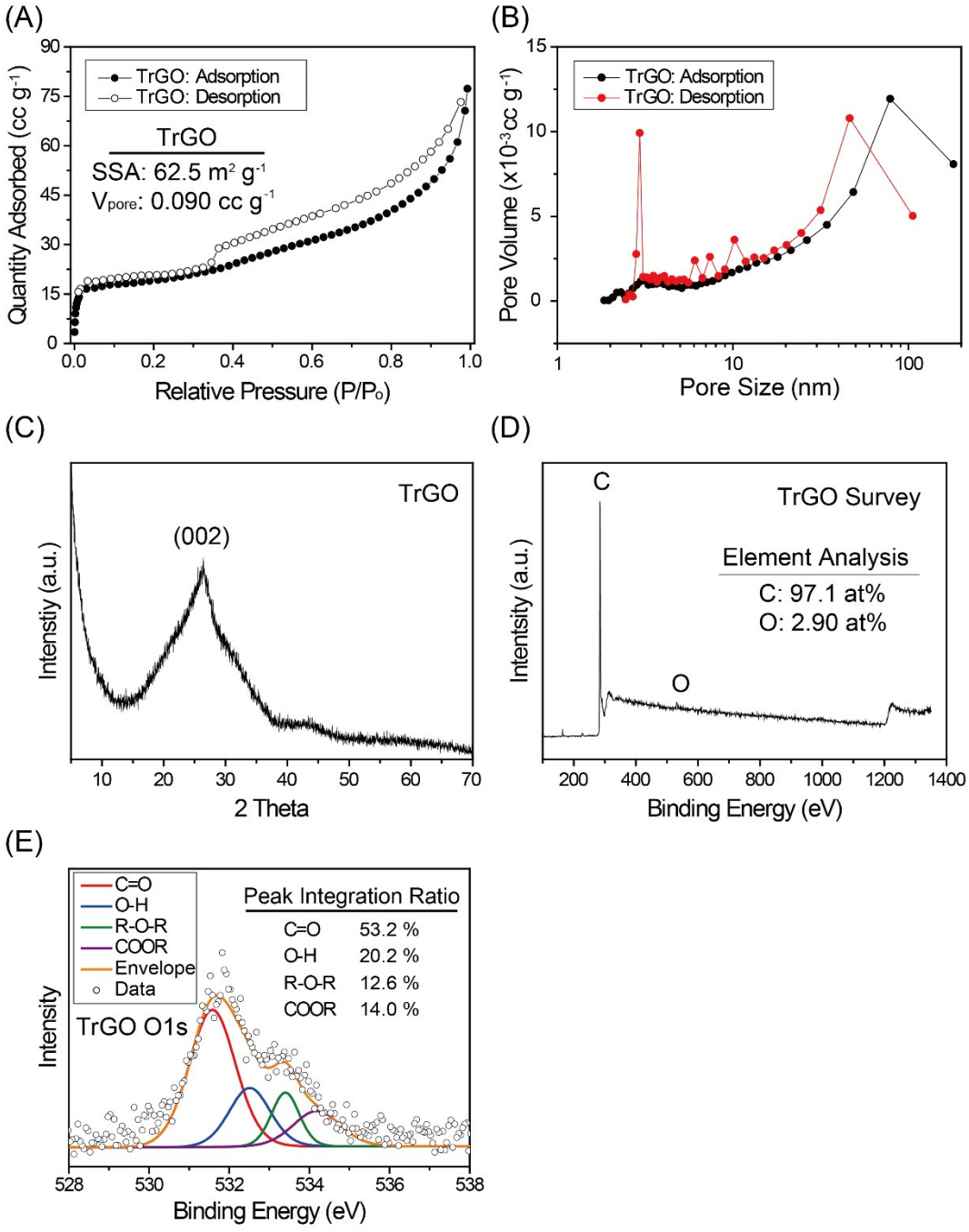

Fig. S3 (A) Ar adsorption/desorption isotherms of TrGO at 87 K. (B) Incremental pore size distribution curves of TrGO from the adsorption (black) and desorption (red) curves using the BJH model. (C) X-ray diffraction pattern of TrGO. (D) XPS survey spectrum of TrGO. The inset indicates the elemental contents from the survey spectrum. (E) XPS O1s spectrum of TrGO. The inset indicates the peak integration ratio of functional groups.

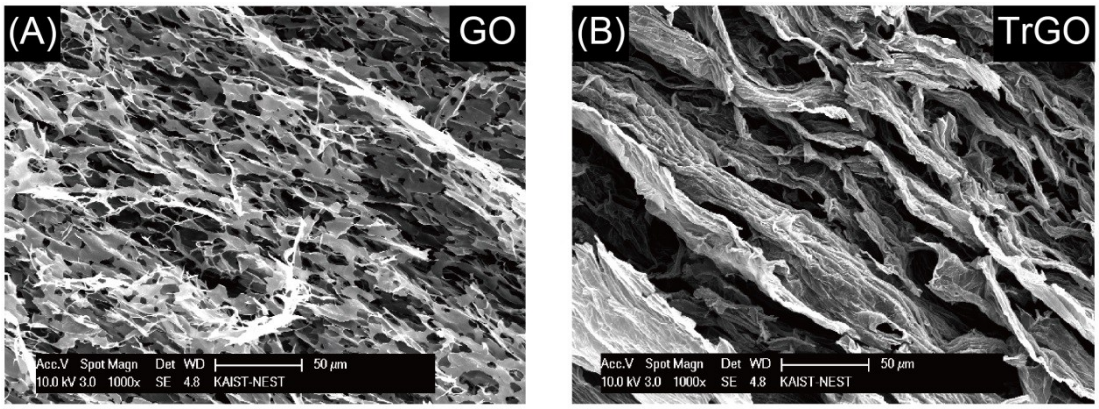

Fig. S4 SEM images of (A) GO and (B) TrGO.

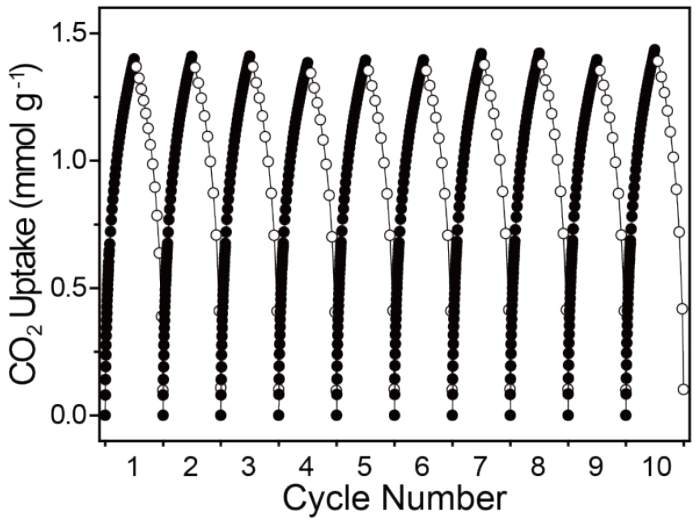

Fig. S5 Repeated CO_2 adsorption/desorption for 10 cycles at 298 K.

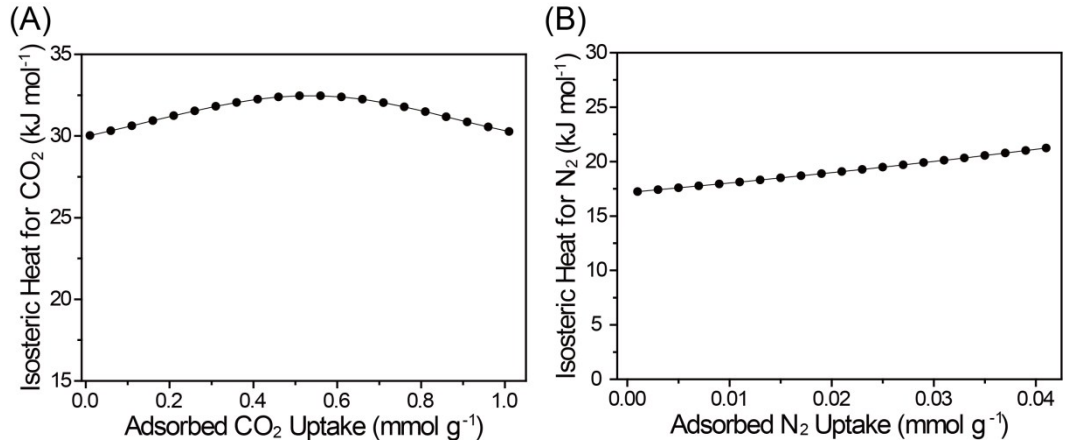

Fig. S6 Isosteric heats (Q_{st}) of GO for (A) CO_2 and (B) N_2 adsorption.

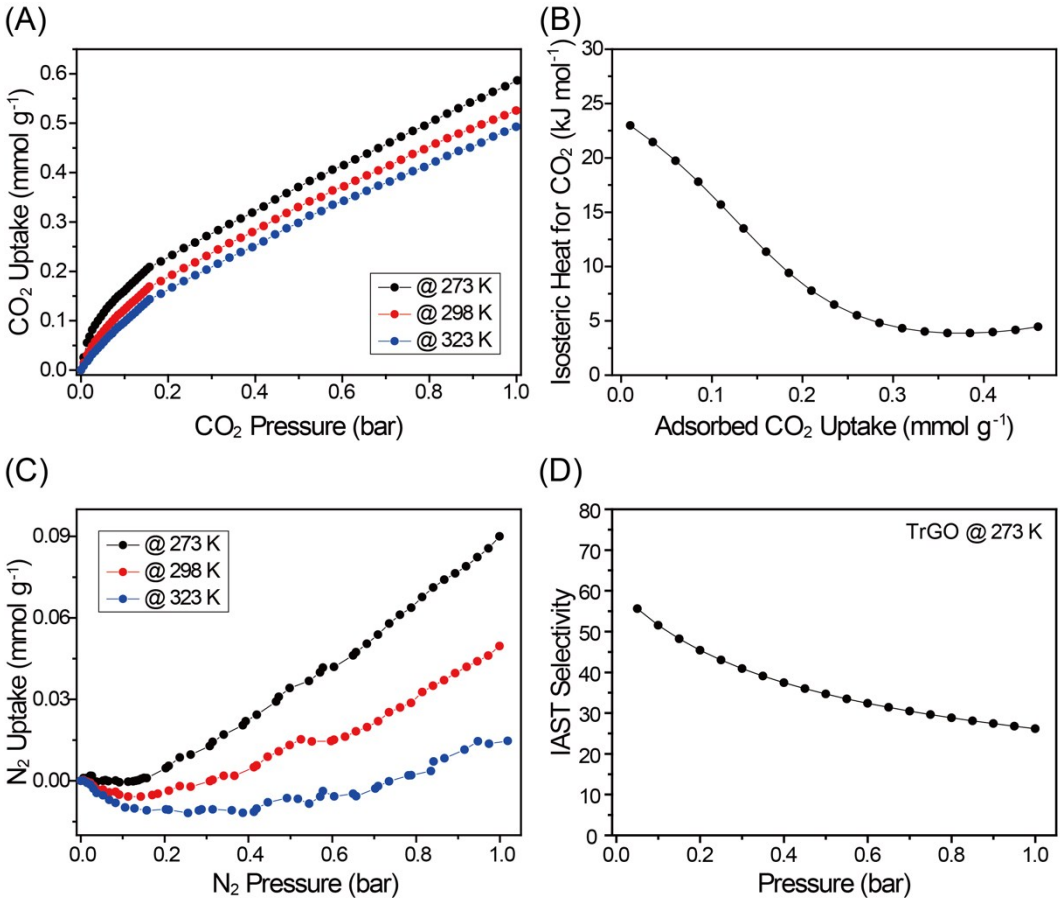

Fig. S7 (A) CO_2 uptake isotherms of TrGO at 273, 298, and 323 K. (B) Isosteric heats (Q_{st}) of TrGO for CO_2 adsorption. (C) N_2 uptake isotherms of TrGO at 273, 298, and 323 K. (D) The calculated IAST CO_2/N_2 selectivity of TrGO for CO_2/N_2 mixture with a molar ratio of 15/85 at 273 K.

The overall N_2 uptake profiles at three different temperatures show negative slopes at the initial N_2 loading, which is attributed to the negligible interaction between N_2 molecules and the TrGO sheets. As a result, the reliable Q_{st} values for N_2 adsorption and IAST selectivity at 298 and 323 K were unable to be obtained.

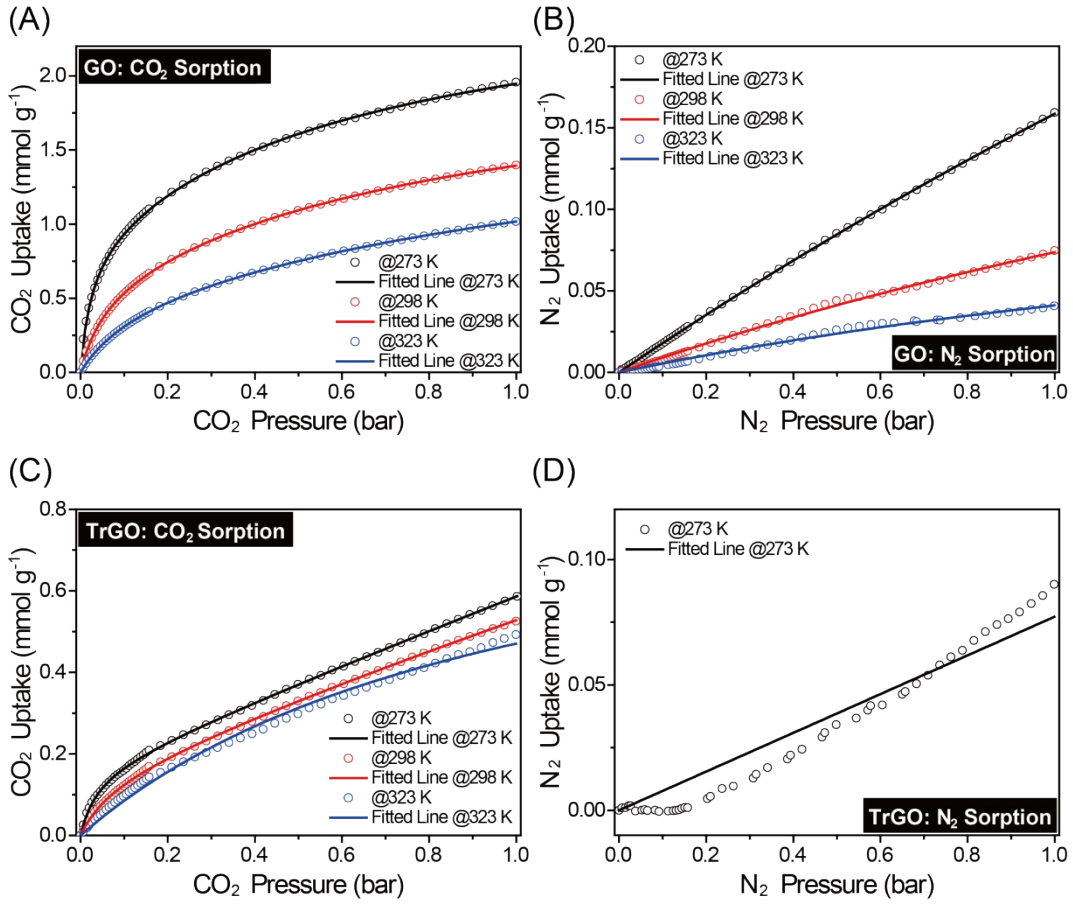

Fig. S8 Langmuir model fitting results for CO_2 and N_2 adsorption of both samples at various temperatures. The detailed parameters are presented in the experimental section and tabulated in Table S1 and S2.

In the case of the TrGO at 298 and 323 K, the reliable fitting of N_2 adsorption was not achieved because of its negligible interaction with N_2.

Table S1. Dual-site Langmuir fitting parameters for CO_2 adsorption.

Samples		$q_{sat,(A)}$ (mmol g^{-1})	$b_{(A)}$ (bar^{-1})	$q_{sat,(B)}$ (mmol g^{-1})	$b_{(B)}$ (bar^{-1})	R^2
GO	@273 K	1.86056	1.46429	0.86188	41.38938	0.99980
	@298 K	1.62785	1.07546	0.58181	18.16883	0.99996
	@323 K	1.56364	0.60963	0.47603	8.46799	0.99999
TrGO	@273 K	27.72184	0.01554	0.16851	25.17124	0.99982
	@298 K	12.62856	0.14717	5.38538	0.14717	0.99986
	@323 K	0.47549	0.9797	0.47549	0.9797	0.99407

Table S2. Dual-site Langmuir fitting parameters for N_2 adsorption.

Samples		$q_{sat,(A)}$ (mmol g^{-1})	$b_{(A)}$ (bar^{-1})	$q_{sat,(B)}$ (mmol g^{-1})	$b_{(B)}$ (bar^{-1})	R^2
GO	@273 K	0.63729	0.14217	0.63729	0.14217	0.99994
	@298 K	0.17649	0.26393	0.17649	0.26393	0.99779
	@323 K	0.07587	0.37123	0.07587	0.37123	0.99085
TrGO	@273 K	108.1800	0.000357	108.1800	0.000357	0.93258
	@298 K[a]	N/A	N/A	N/A	N/A	N/A
	@323 K[a]	N/A	N/A	N/A	N/A	N/A

a. Due to the negligible interaction between TrGO and N_2 gas.