

Electronic Supplementary Information (ESI)

**Thermodynamically Favorable Route to the Synthesis of Nanoporous Graphene
Templated on CaO *via* Chemical Vapor Deposition**

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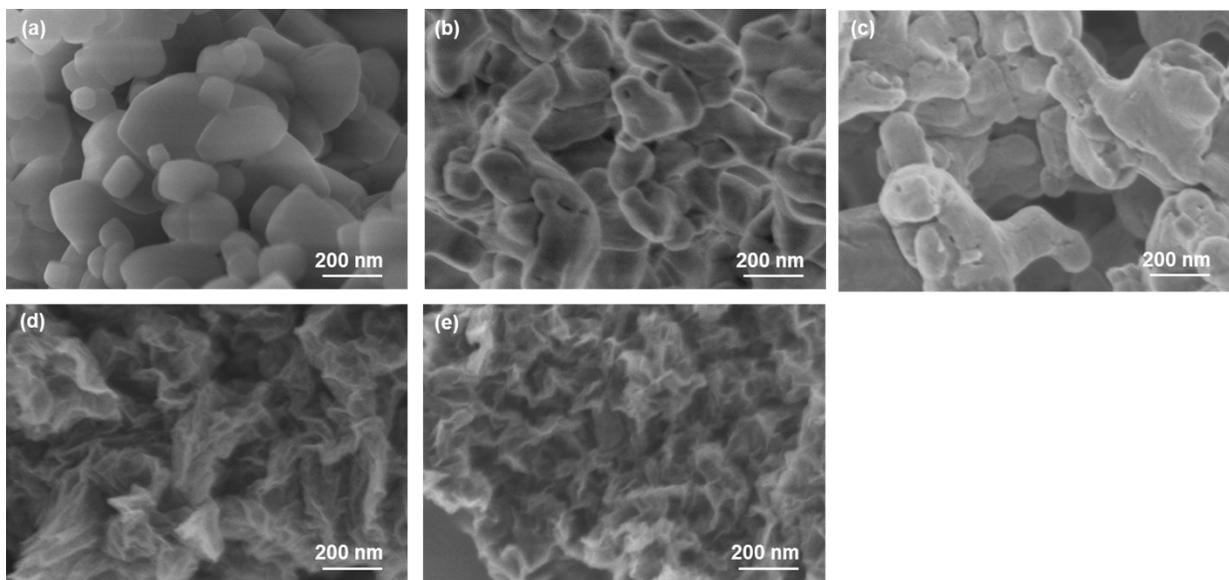


Fig.S1 SEM images of (a) SK-I, (b) SK-I_900, (c) C/SK-I, (d) TC_SK-I, and (e) TC_SK-I_A

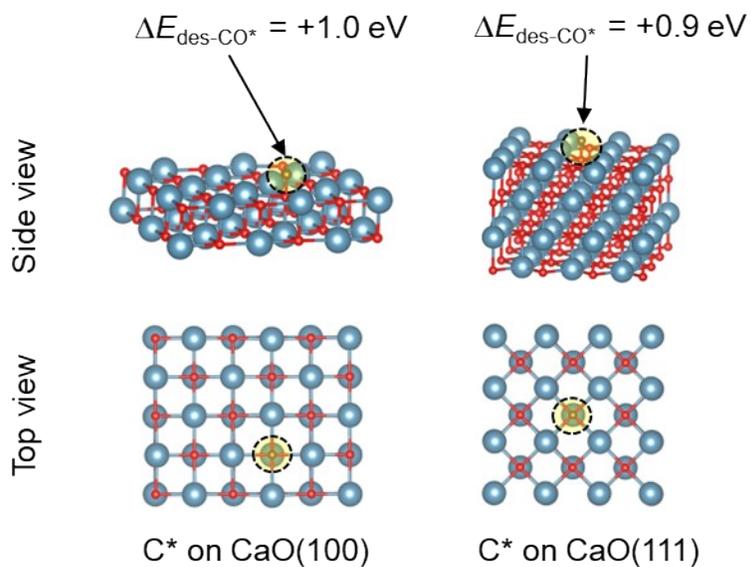


Fig. S2 Side and top views of the optimized structural models of (left) C* on CaO(100), , and (right) C* on CaO(111) with the values of the CO desorption energy ($\Delta E_{\text{des-CO}^*}$) from the labeled C* on O sites. The blue, red, and brown balls represent Ca, O, C atoms, respectively.

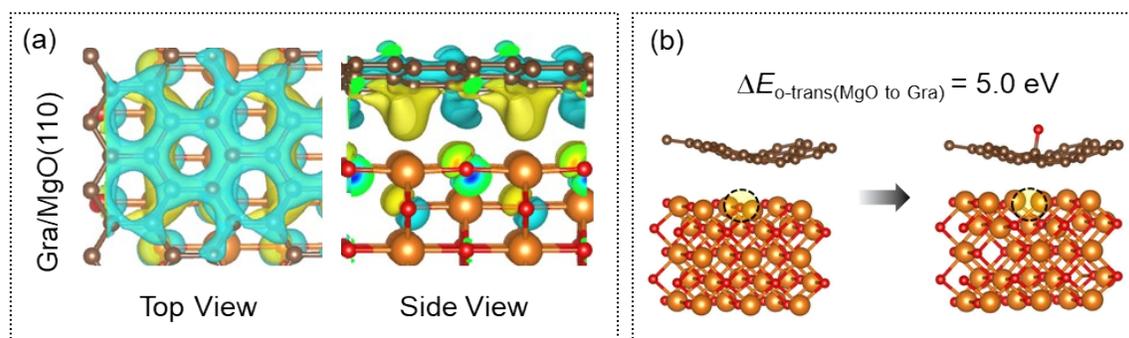


Fig. S3 (a) Charge density mappings of graphene/MgO(110) (Gra/MgO(110)). (b) The optimized structural model of Gra/MgO(110) and O* on Gra/MgO(110) with the energy required for transferring O from MgO to the surface graphene ($\Delta E_{O\text{-trans(MgO to Gra)}}$). The red, orange, and brown balls represent O, Mg, and C atoms, respectively.

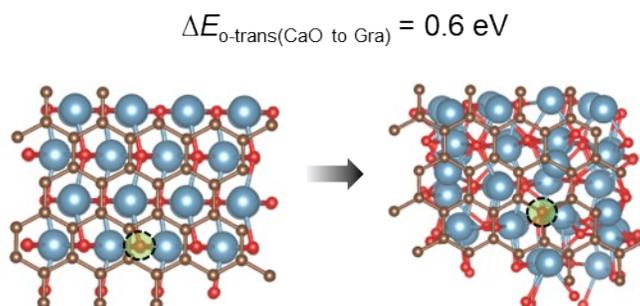


Fig. S4 Top views of the optimized structural models of Gra/CaO(110) and O* on Gra/CaO(110) with the energy required for transferring O from CaO to the surface graphene ($\Delta E_{O\text{-trans(CaO to Gra)}}$). The green region locates the transferred O.

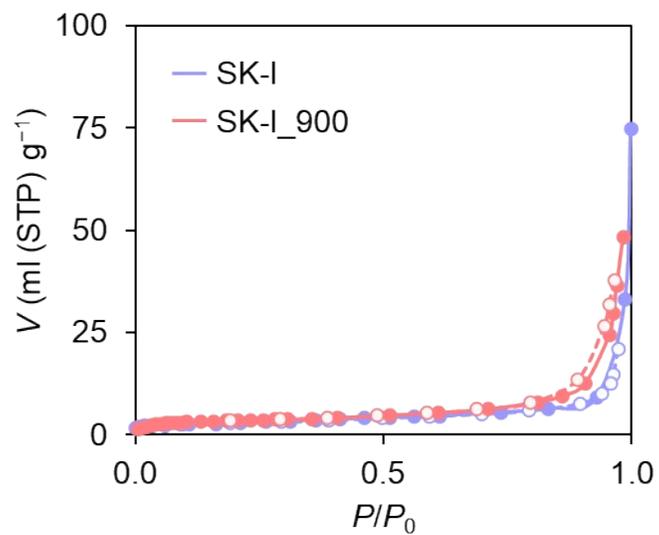


Fig. S5 N_2 adsorption/desorption isotherms at $-196\text{ }^\circ\text{C}$ for SK-I and SK-I_900.

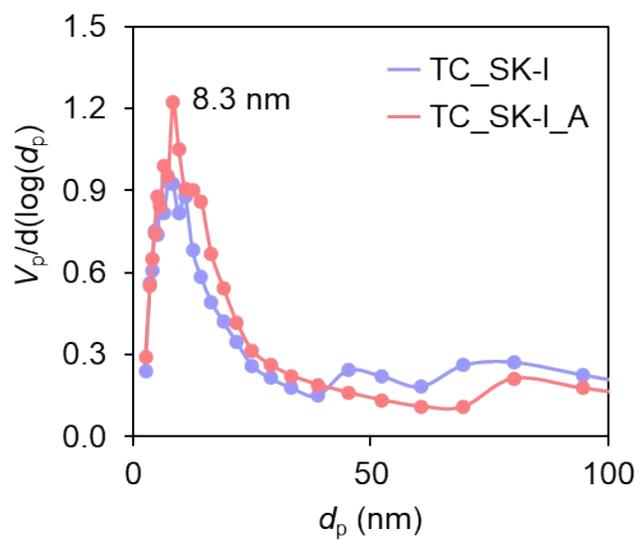


Fig. S6 Pore-size distributions calculated by the BJH method for TC_SK-I and TC_SK-I_A.

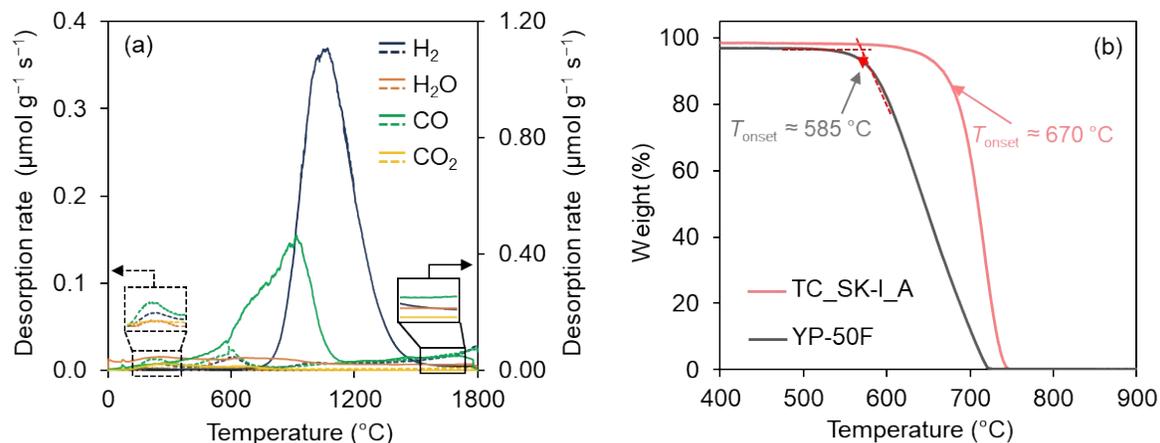


Fig. S7 (a) Gas evolution patterns of YP-50F (solid lines) and TC_SK-I_A (dashed lines) during the high-sensitivity TPD measurement up to 1800 °C. (b) The oxidation resistance of YP-50F and TC_SK-I_A characterized by TG measurement in air. The heating rate is 5 °C min⁻¹.

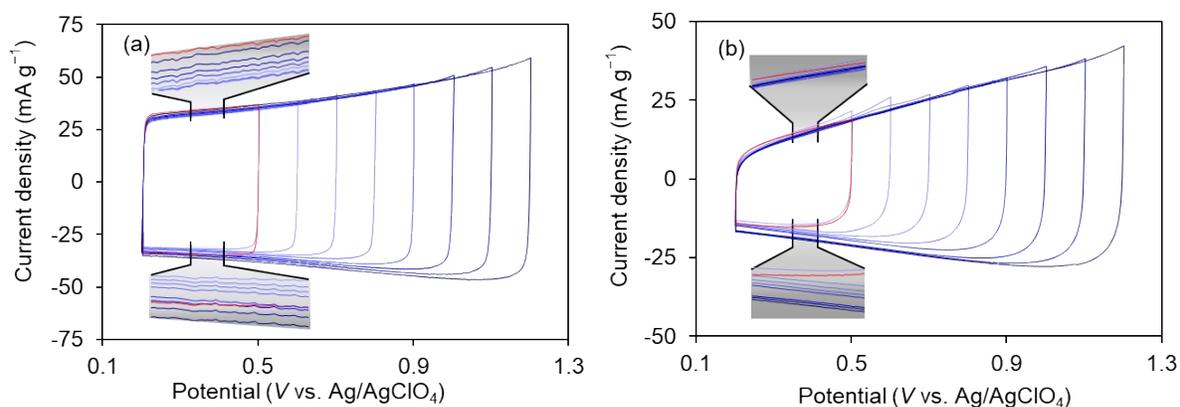


Fig. S8 Cyclic voltammograms measured with a scan rate of 1 mV s⁻¹ using (a) TC_SK-I and (b) TC_SK-I_A. A three-electrode cell was used with 1 M Et₄NBF₄/propylene carbonate at 25 °C. The blue lines represent the data during the upper limit potential inclining step in the range of 0.5-1.2 V. The red line is the data measured between 0.2 and 0.5 V following the CV performed from 0.2 to 1.2 V.

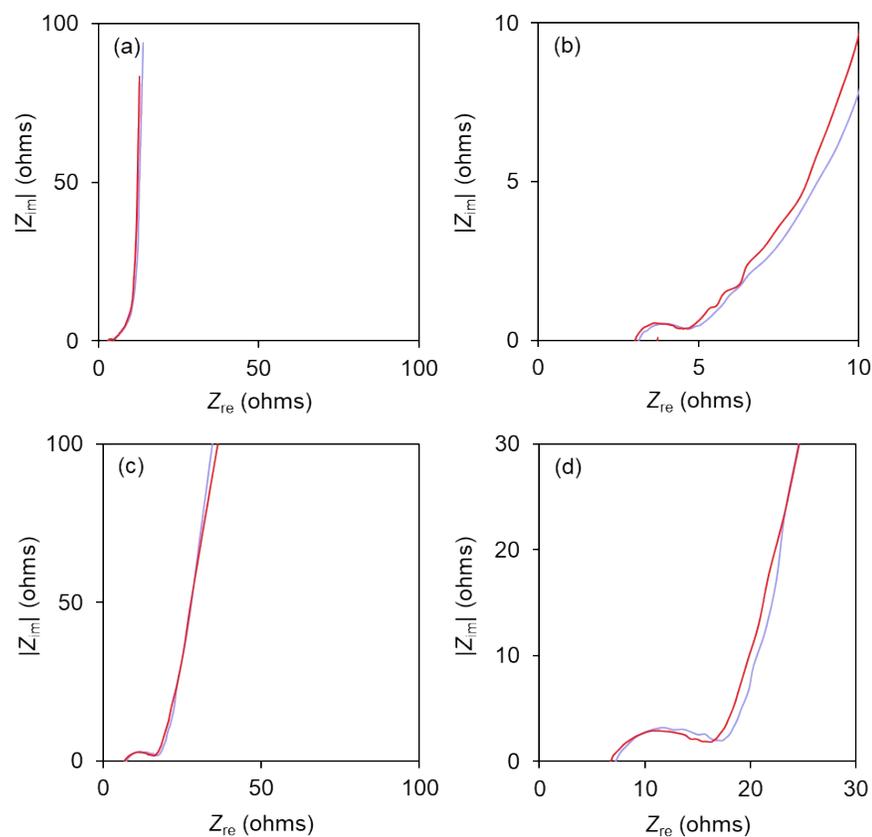


Fig. S9 The Nyquist plots of TC_SK-I and TC_SK-I_A measured by using a three-electrode cell in 1 M Et_4NBF_4 /propylene carbonate at 25 °C. Nyquist plots at OCV (0.2 V) obtained throughout cyclic voltammetry (CV) measurement of (a,b) TC_SK-I (c,d) TC_SK-I_A. The blue line represents the data before the CV measurement. The red line is data measured following the CV performed from 0.2 to 1.2 V.

Table S1 Impurities contained in CaCO₃ analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES). The data are kindly provided by Shiraishi Central Laboratories Co., Ltd.

Sample	Impurity content (ppm)					
	Na	Mg	Fe	Sr	Si	Al
SK-I	27.8	4.2	3.5	3.5	6.9	<0.1