Supporting Information

Convert CO₂ into Oxygenated Alkynyl Carbon Material with High Electrochemical Performance through Mechanochemical Reaction with CaC₂

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Experimental Section

Mechanochemical reaction process of CaC₂ and CO₂

Table S1 lists the detailed data of the mechanochemical reaction process. For this mechanochemical reaction, a planetary ball mill with four 250 mL 304-stainless steel pots that contains approximately 350 g 304-stainless steel balls with three different diameters (175 g-5 mm, 105 g-8 mm, and 70 g-10 mm) was used. The detailed composition of CaC₂ used in this experiment is 75 wt% of CaC₂, 22 wt% of CaO, and 3 wt% of other impurities including CaSiO₃, Ca₃P₂, Ca₈, Fe₂O₃, Al₂O₃, etc. Before reaction, the massive CaC₂ was pulverized into powder by a high speed crusher, then the CaC₂ powder was screened by 100 mesh sieve and standby. The CaC₂ powder was added at once, while the gaseous CO₂ was added in three installments. Before operation, 14.79 g (0.17 mol) of CaC₂ powder (75 wt%) was added into the pot. The pot was subsequently sealed air tight using a vacuum controlled system. For the first round, 0.3 MPa of CO₂ was filled into the pot at 10.6°C, and the sealed pot was fixed to a planetary ball mill. The planetary ball mill was operated at 650 rpm for 120 min under vacuum conditions and ambient temperature, with a cooling interval of 5 min after every 30 min for avoiding excess temperature increase. Then, the pot was purged with high purity N₂ (Figure S1), the purged gas was passed into clear limewater, and the limewater does not become turbid. After that, the pot was cooled down and opened to check the reaction. For the second round, 0.3 MPa of CO₂ was filled into the pot at 12.8°C, and the planetary ball mill was operated at 650 rpm for 120 min. For the third round, 0.3 MPa of CO_2 was filled into the pot at 15.2°C, and the planetary ball mill was operated at 650 rpm for 120 min. Again, the pot was purged with high purity N₂ (Figure S1), the purged gas was passed into clear limewater, and the limewater does not become turbid. Then, the milled solid was sampled and further investigated by XRD and FT-IR. The milled solid was immersed in a dilute acid (HCl, CH₃COOH, or HNO₃) for a period of time, filtered using a microfiltration membrane, and washed three times by ultrapure water. Finally, 5.09 g OACM was obtained after vacuum drying at 120 °C for 5 h. In addition, to verify the influence of CaO, we

supplemented the mechanochemical reaction of CaO and CO₂, and the result showed they did not react under the same experimental conditions. So, the influence of CaO was excluded. Besides, control experiment was made: equal amounts of CaC₂ was taken, and treated in the same manner under Ar atmosphere (0.3 MPa) but in the absence of CO₂. It shows that the pressure was not released, and the CaC₂ powder keeps in gray after milling. These indicate no reaction occured when only CaC₂ was milled in the absence of CO₂. At the same time, the amount of the insoluble impurities from CaC_2 was obtained.

Table S1. Detailed data for the mechanochemical reaction process.									
Experimental Condition	CaC ₂ Input	CO ₂ Input					Sample	0.4.634	
		First Round Second Round		Round	Third Round		After	Viald	
		Input	Temp.	Input	Temp.	Input	Temp.	Reaction	Ticiu
650 rpm; 250 mL pot; 350 g balls	14.79 g 0.17 mol	0.3 MPa	10.6 °C	0.3 MPa	12.8 °C	0.3 MPa	15.2 °C	0.54 g	5.09 g



Figure S1. The tail gas examining for the after milling reaction system.

Furthermore, the synthesis process was repeted, unreacted CaC_2 after milling was hydrolyzed, and the volume of the resultant C_2H_2 was measured. Then, the carbon balance analysis for the process was conducted. As shown in **Table S2**, the results show that the carbon contribution of CaC_2 for the resultant OACM is about 77.5% [(4.18-1.45)/3.52], and the carbon in CO_2 is the sole source for the residual carbon of OACM, which directly reveals that the resulting OACM contains not only carbon originated from CaC_2 but also carbon from CO_2 .

Table S2. Carbon balance analysis for the mechanochemical reaction process.					
	C-related Materials	Dosage	C-content (g)	Total (g)	
Innut C	CaC ₂ (75wt% purity)	14.88 g	4.18	5.07	
Input-C	CO_2	3.27 g	0.89	5.07	
Output C	OACM (74.5wt% C content)	4.73 g	3.52	4.07	
Output-C	C_2H_2	1350 mL	1.45	4.97	

Furthermore, **Table S3** lists the possible reaction circumstance and correlation analysis. For series 1, the practical OACM yield is 93.9%, but the formed alkynyl-linked intermediates, $[O-C-O]^{2-}$, are generally unstable and easily rearranged to carbonyl (C=O). As to series 2, the practical OACM yield is 120.3%, but the remained carbonyl provide incentive for the reaction proceeding due to the high carboanionic and nucleophilic nature of $[C=C]^{2-}$ from CaC₂. Correspondingly, the structural formula in series 3 shows that no additional easily attacked positively charged C-atom left in the resultant OACM, and thus the reaction can be terminated. Thus, for the reaction of CaC₂ and CO₂, series 3 is the most practical posibility as evidenced by the very high consistency between theoretic and experimental values in element content and OACM yield (99.4%). Meanwhile, this also reveals that the reaction reached deep level, indicating its high efficiency under mechanochemical treatment.

Tal	Table S3. Possible reaction circumstance and correlation analysis.							
Structural No. Formula of		Reaction Equation	OACM	Theoretic Element Content (wt%)		Theoretic OACM Vield (g)	Practical OACM	
UA	OACM			С	0	r leiu (g)	1 ieiu (70)	
1	$\begin{array}{c} \sigma & \sigma & \sigma \\ - c - c = c - c - c = c - c - c - c \\ \sigma & \sigma & \sigma \end{array}$	$CaC_2+CO_2=$ [C ₃ O ₂] ²⁻ +Ca ²⁺	C_3O_2	52.9	74.1	5.42	93.9	
2	0 0 0 −c−c≡c−c−c≡c−c−	$CaC_2+CO_2=$ C_3O+CaO	C ₃ O	69.2	30.8	4.23	120.3	
3		$3CaC_{2}+2CO_{2}=$ $2[C_{4}O]^{-+}2CaO$ $+Ca^{2+}$	C ₄ O	75.0	25.0	5.12	99.4	

Besides, a brief techno-economic analysis for this process was made. As shown in **Table S4**, the rough overall consumption (including raw materials consumption, energy consumption and auxiliary materials consumption) is about \pm 55 for per kilogram of OACM generated. This reveal the good technical and economic feasibility for the mechanochemical synthesis strategy of OACM.

Table S4. Techno-economic analysis for mechanochemical synthesis of OACM.						
	Materials	Dosage	Unit price (Y)	Total (¥)		
Raw materials	CaC ₂	~3 kg	~3	. 0		
consumption	CO_2	~0.7 kg		~9		
Enorgy consumption	Ball milling	$\sim \!\! 40 \text{ kW h}$	0.61~0.65	22		
Energy consumption	Drying	$\sim \! 10 \ kW \ h$	0.61~0.65	~55		
Auxiliary materials	H ₂ O	~3 kg	4.1	12		
consumption	Acid (HCl)	~3.5 kg	~0.35	~13		
Note: data in the table are calculated per kilogram of OACM generated.						

In addition, CO_2 in CaC_2 industry can be internal balance and cycle based on the the following reaction processes. So no excess CO_2 releases for the CaC_2 industrial chain in theory. Thus, the CO_2 consumed in this mechanochemical process can be deemed as the net CO_2 consumption from the global environment. Based on these, this mechanochemical process consumes about 0.7 kg of CO_2 for per kilogram of OACM generated, which highlights its high environmental value. Thus, this mechanochemical strategy provides an innovative approach for high value resource utilization of CO_2 , and the implementation of this process will benefit to the mitigation of CO_2 . In the background of the increasing energy consumption and greenhouse gases emission, we hope this strategy can make contributions for tackling climate change and global warming.

$$CaCO_{3} \rightarrow CaO + CO_{2}$$

$$CaO + 3C \rightarrow CaC_{2} + CO$$

$$CaC_{2} + 2H_{2}O \rightarrow Ca(OH)_{2} + C_{2}H_{2}$$

$$Ca(OH)_{2} + CO_{2} \rightarrow CaCO_{3} + H_{2}O$$

S5

Characterization instruments

X-ray diffraction (XRD) was conducted using a Bruker D8 Advance XRD system (BRUKER, Germany) with Cu K α radiation at a speed of 8° min⁻¹ in the 2 θ range of 5°-85°. Fourier transform infrared (FT-IR) was conducted using Nicolet 6700 (ThermoFisher Scientific, US) with the KBr disk method from 400 cm⁻¹ to 4000 cm⁻¹. Elemental analysis was conducted using a FlashSmart analyzer (Thermo Fisher, US). Scanning electron microscopy (SEM) images were recorded on a NANO SEM430 microscope (ThermoFisher Scientific, US). Transmission electron microscopy (TEM) images were recorded on a JEM-2100 microscope (Hitachi, Japan). For TEM measurements, the sample was suspended in ethanol and transferred onto a microgrid. The X-ray energy-dispersive spectroscopy (EDS) with elemental mapping was recorded on an X-MaxN 80T system (OXFORD Instruments, UK) attached to the TEM. N₂ adsorption-desorption isotherm analysis was performed at 77 K with a computer-controlled nitrogen gas adsorption analyzer (Micromeritics, US), and the Brunauer-Emmett-Teller (BET) and density functional theory (DFT) methods were employed for the determination of specific surface area and pore size distribution, respectively. Raman spectrum was recorded on a Raman microscope (Renishaw, UK) under an excitation of 532 nm. Solid-state ¹³C NMR spectrum was conducted on 400WB AVANCE III (Bruker, Switzerland). Thermogravimetric analysis and differential scanning calorimetry (TG-DSC) was performed with a TGA/DSC3+HT/1600 thermal analyzer (Mettler Toledo, Switzerland). The TG-DSC measurement was conducted at heating rate 10°C min⁻¹ and carried out under argon atmosphere with a flow rate of 50 mL min⁻¹ in the temperature range from 100 to 1000°C. X-ray photoelectron spectroscopy (XPS) was recorded on a Thermo Escalab 250Xi instrument (Thermo Electron, US).

Electrochemical performance measurements

The working electrodes were fabricated by mixing the OACM and polytetrafluoroethylene (PTFE) in a weight ratio of 8:1 in ethanol. Notably, no conductive filler (e.g., acetylene black) was used because of its high conductivity. Then, the mixture was rolled into sheets and pressed on nickel foam current collector with a coating area of 1 cm². Finally, the as-fabricated electrode was dried at 120 °C for 10 h in a vacuum oven for completely removing ethanol. Dry electrode was assembled as symmetric supercapacitors in a three-electrode cell with platinum foil and Hg/HgO electrodes as the counter and reference electrode, respectively. A 6 M KOH solution was used as the electrolyte.

CV, GCD and EIS investigations were conducted on a CHI 660E electrochemical workstation (Chenhua Instrument, Shanghai, China). CV tests were investigated between -1.0 and 0 V (vs. Hg/HgO) at scan rates ranging from 1 to 100 mV s⁻¹. GCD test was conducted at the same potential range at current densities ranging from 0.15 to 15.0 A g⁻¹. EISs were characterized at open-circuit potential in the frequency range from 100 kHz to 0.01 Hz with amplitude of 5 mV. The long cycling performance was conducted by GCD method under 3.0 A g⁻¹ at the same potential range on a LANHE supercapacitance test system (LAND Electronics Co., Ltd., Wuhan, China).

The gravimetric specific capacitance $(C_m, F g^{-1})$ of the OACM electrode was calculated using CV and GCD curves. On the basis of the CV curves, the C_m value of the OACM electrode at various scan rates was calculated as follows:

$$C_{\rm m} = \frac{\int I dV}{2m\nu\Delta V}$$

Here, m is the mass of the OACM (g), v is the potential scan rate (V s⁻¹), Δ V is the range of potential (V), I is the response current (A), and \int IdV is the mathematical integral of the CV curve. Based on the GCD curves, the C_m value of the OACM electrode at various current densities was calculated as follows:

$$C_{\rm m} = \frac{I\Delta t}{m\Delta V}$$

Here, m is the mass of the OACM (g), I is the charge/discharge current (A), ΔV is the potential range of charge/discharge (V), and Δt is the discharge time (s).

The relaxation time (τ_0, s) of the OACM electrode was calculated from Bode plot as follows:

$\tau_0 = 1/f_0$

Here, f_0 is the characteristic frequency of OACM at the phase angle of -45°.

Besides, electrical conductivity of the obtained OACM was measured using AC 4-probe method on a RTS-9 dual electric four-probe tester (4-Probe Technology Ltd, Guangzhou, China). Before test, the power was pressed to sheet with a pressure of 10 MPa.



Figure S2. The SEM images of OACMs at different magnifications.

$S_{BET} (m^2 g^{-1})$	$S_{mic} (m^2 g^{-1})$	V_{total} (cm ³ g ⁻¹)	$V_{mic} (cm^3 g^{-1})$	V _{mes} (cm ³ g ⁻¹)	D _{ap} (nm)
687	265	0.73	0.14	0.59	2.7

Table S5. Detailed results from the N₂ adsorption-desorption analysis.

 $S_{\text{BET}}\!\!:$ specific surface area obtained by the BET method;

S_{mic}: micropore specific surface area;

V_{total}: total pore volume;

V_{mic}: micropore volume;

V_{mes}: mesopore volume;

D_{ap}: average pore diameter.



Figure S3. Additional TEM and HRTEM images of OACM.

Tashualagiaal maang			Eleme	nt Cont	ent in C	ACM/w	vt%		
l echnological means	С	0	Н	S	Ν	Ca	Fe	Si	Σ
Elemental Analysis	74.5	18.9	1.0	0.8	0.2	/	/	/	95.4
EDS	81.0	19.0	/	/	/	/	/	/	100.0
XPS	72.9	26.0	/	/	/	0.5	0.2	0.4	100.0

Table S6. Compositional results obtained from EA, XPS, and EDS.



Figure S4. The water contact angle measurement for OACM.



Figure S5. The TG-DSC analysis of OACM in argon atmospheres.



Figure S6. XPS survey spectrum of OACM (the element contents listed in Table S6).

Element	Carbon bond	Position (eV)	FWHM	Area ratio (%)
	$C-C(sp^2)$	283.5	2.8	13.1
	C–C (<i>sp</i>)	284.8	1.6	43.1
C 1s	$C-C(sp^3)$	285.5	2.6	15.8
	С–О	286.1	3.0	12.8
	C=O	288.6	1.8	15.3
	C=O	531.0	3.6	16.2
O 1s	С-ОН/С-О-С	532.3	2.8	80.7
	O–C=O	533.5	1.1	3.1

Table S7. The fitting results of the XPS C 1s and O 1s narrow scan.



Figure S7. CV curves of the OACM electrode at different scan rates ranging from 25 to 100 mV s⁻¹ in the potential range between -1.0 and 0 V (vs. Hg/HgO). The C_m values of the OACM electrode calculated using the above CV curves are listed in **Table S8**.



Figure S8. GCD curves of the OACM electrode at different current densities ranging from 2.25 to 15.0 A g^{-1} in the potential range between -1.0 and 0 V. The C_m values of the OACM electrode calculated using the above GCD curves are listed in **Table S8**.

Methods and expen	rimental conditions	C _m (F g ⁻¹)
	0.15	121.8
	0.30	110.7
	0.75	101.6
GCD curves at	1.50	97.5
different current	2.25	92.5
densities (A g ⁻¹)	3.00	90.9
	7.50	82.5
	15.0	75.0
	1	104.1
	2	102.7
	5	95.7
CV curves at different	10	90.8
scan rates (mV s ⁻¹)	25	83.9
	50	77.9
	75	74.0
	100	70.6

Table S8. C_m values of the OACM electrode obtained from CV and GCD curves.

	G • @	Capacitan		
Carbon materials	Specific surface area	elec	Cost	
	(m² g ²)	C _m (F g ⁻¹)	C _v (F cm ⁻³)	_
Carbon nanotubes	120–500	50-100	<60	High
Graphene	2630	100-205		High
Commercial activated carbon	1000–3500	<200	<80	Low
Activated carbon fibers	1000–3000	120-370	<150	Medium
Carbon aerogels	400–1000	100-125	<80	Low
Graphyne	330–720	48–135		Medium
Graphdiyne	600–1300	<200		High
Naphyne	970.9	154	158.6	Low
OACM (this study)	686.9	121.8	166.6	Low

Table S9. Comparison of various carbon electrode materials for supercapacitors.

C_m: gravimetric specific capacitance

C_v: volumetric specific capacitance

Carbon materials	Electrical conductivity (S m ⁻¹)	Ref.
Carbon black	100~104	S18-20
CNT fibers	700~3000	S21-22
CNT-graphene hybrid fibers	1200	S21
Graphene fibers	3500	S21
Graphene aerogel	87	S23
Graphene oxide papers	1350~2100	S24
Reduced graphene oxide	1~1500	S25
Activated carbon	1~2000	S26
Onion like carbons	400	S27
Graphdiyne nanowires	1900	S28-29
γ-Graphyne	222~4070	S30-31
Naphyne	1490	S15
OACM	1522	This work

 Table S10. The electrical conductivity performance of various carbon materials.



Figure S9. The electrochemical performance comparison of the OACMs obtained from different mechanochemical conditions.

Preliminary results show that the OACM can be efficiently synthesized under milder mechanochemical condition (rotate speed of 550 rpm and milling time of 60 min) compared with that of original used (rotate speed of 650 rpm and milling time of 120 min). Furthermore, electrochemical investigation for the resultant OACM under milder condition shows an improved performance. As shown in Figure S9a, the GCD curve of new OACM electrode also exhibit symmetrical triangular shape with extremely small IR drop, which is almost unchangeable compared with that of original OACM, demonstrating its good capacitive character with highly reversible systems and small internal resistance. Meanwhile, its GCD curve shows the new OACM has higher C_m value (112.7 F g^-1) than the original one (97.5 F g^-1) under same current density (1.5 A g⁻¹). In addition, the Nyquist plot of new OACM (Figure **S9b**) exhibits lower equivalent series resistance (0.48 Ω), demonstrating its better conductivity. The improved electrochemical performance may be attributed to avoiding the excessive mechanical disruption for the porous structure of the resultant OACM during the reaction. This result indicates the mechanochemical synthesis condition has great influence on its electrochemical performance, deserving constant optimization and in-depth study.

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