# Supporting information

Effective combination catalyst of  $CeO_2$  and zeolite for direct synthesis of diethyl carbonate from  $CO_2$  and ethanol with 2,2-diethoxypropane as a dehydrating agent

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## Materials

All catalysts and substrates were purchased or supplied from chemical companies. The details are shown below. Before the activity test,  $CeO_2$ ,  $Al_2O_3$ ,  $Nb_2O_5$ ,  $ZrO_2$ , and  $Ta_2O_5$  were calcined at 873 K for 3 hours, and  $SiO_2$  and  $SiO_2 \cdot Al_2O_3$  were calcined at 773 K.

Solid catalyst	Company name product name
Al <sub>2</sub> O <sub>3</sub>	Nippon Aerosil, AEROXIDE Alu C
Nb <sub>2</sub> O <sub>5</sub>	Sigma-Aldrich (purity: 99.99 %)
Ta <sub>2</sub> O <sub>5</sub>	FUJIFILM Wako Pure Chemical Corporation (purity: 99.9 %)
$ZrO_2$	Daiichi Kigenso Kagaku Kogyo, RC-100
SiO <sub>2</sub>	Fuji Silysia Chemical LTD., G-6
CeO <sub>2</sub>	Daiichi Kigenso Kagaku Kogyo, CeO2-HS
SiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub>	JGC C&C, LA silica-alumina
H-FAU (SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =25)	Tosoh Corporation, HSZ-371NHA
H-MFI (SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =24)	Tosoh Corporation, HSZ-822HOA
H-*BEA (SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =28)	Tosoh Corporation, HSZ-931HOA
H-MOR (SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =18)	Tosoh Corporation, HSZ-640HOA
H-FAU (SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =5.5)	Tosoh Corporation, HSZ-320HOA

Table S1 (a) List of solid catalysts

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Reagent and substrate	Company name	Purity /%
HNO <sub>3</sub>	FUJIFILM Wako Pure Chemical Corporation	69~70
$H_2SO_4$	FUJIFILM Wako Pure Chemical Corporation	95.0
H <sub>3</sub> PO <sub>4</sub>	Nacalai Tesque	85
Benzenesulfonic acid monohydrate	Tokyo Chemical Industry	>98.0
Maleic acid	Kanto Chemical	>99.0
Pyridine-2-carboxylic acid	Tokyo Chemical Industry	>99.0
Benzoic acid	FUJIFILM Wako Pure Chemical Corporation	>99.5
2,2-Diethoxypropane	Tokyo Chemical Industry	>95.0
Ethanol	Kanto Chemical	>99.5
2,2-Dimethoxypropane	Tokyo Chemical Industry	>98.0
Methanol	Kanto Chemical	>99.5
1-Hexanol	Tokyo Chemical Industry	>98.0

#### Activity test

The typical reaction procedure for DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> and an acid catalyst is as follows: Reactions were carried out in autoclaves with 190 ml inner volume. A magnetic stirrer, CeO<sub>2</sub> 0.5 g, an acid catalyst 0.01 g, EtOH 140 mmol, and 2,2-diethoxypropane 21 mmol were added into the autoclave in order. The autoclave was sealed up and purged by 1 MPa (r.t.) CO<sub>2</sub> two times, and then pressured to 5 MPa (r.t.) by CO<sub>2</sub>. The autoclave was heated to 393 K, and the time when the autoclave reached 393 K was defined as beginning of the reaction (0 h). The CO<sub>2</sub> pressure reached 6.5 MPa at 393 K. After reaction, the autoclave was cooled down to room temperature by a water bath. The reaction mixture was recovered with 1-hexanol (0.2 g) as the internal standard and analyzed by GC-FID (TC-WAX). Figure S3 showed the typical GC chart of DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> + H-FAU. Conversion, yield, and balance were calculated by the following equations.

DEP conversion  $/\% = [1 - DEP_{(Residual)}) / DEP_{(Introduced)}] \times 100$ 

DEC yield (on DEP basis)/% = DEC\_{(detected after reaction)} / DEP\_{(Introduced)} \times 100

When the DEP conversion was below 20 %, the DEP conversion was calculated by the following equation.

 $DEP \ conversion \ /\% = [Acetone_{(detected after reaction)} + 2-Ethoxypropene_{(detected after reaction)} + By-products_{(acetone-derived)}] / [Acetone_{(detected after reaction)} + 2-Ethoxypropene_{(detected after reaction)} + DEP_{(residual)} + By-products_{(acetone-derived)}] \times 100$ 

DEP balance /% = [Acetone<sub>(detected after reaction)</sub> + 2-Ethoxypropene<sub>(detected after reaction)</sub> + Byproducts<sub>(acetone-derived</sub> + DEP<sub>(Residual)</sub>] / DEP<sub>(Introduced)</sub> × 100

#### **Measurement of reaction rates**

The typical procedure for measurement of the reaction rates is as follows: A magnetic stirrer,  $CeO_2 0.03$  g, an acid catalyst 0.01 g, EtOH 140 mmol, and 2,2-diethoxypropane 21 mmol were put into the autoclave reactor with 190 ml inner volume under air. The autoclave reactor was sealed, and the air content was purged by 1 MPa Ar twice. The autoclave was heated to 393 K, and the autoclave was pressured by  $CO_2$  to 5 MPa at 393 K, and the time was defined as beginning of the reaction (0 h). After the reaction, the autoclave reactor was cooled to room temperature by a water bath. Recovery and analyses of the reaction mixture were the same as the above procedure.

### **Reusability test**

The typical method for reusability test is as follows: Some reactions (2-4 batches) were carried out at the same time, and the reaction operation is the same as the typical procedure described in the activity test. After the reaction, the reaction mixtures were analyzed by GC first, and the reaction mixtures were separated into the liquid phase and solid phase by centrifugation. The liquid phase was removed, and ethanol was added into the collected solid and well-mixed for washing. The obtained solid solutions were separated by centrifugation again. The washing operation of the collected solid with ethanol was repeated at least 5 times. The obtained wet solids were dried at 383 K overnight and calcined at 623 K for 3 hours (The catalyst loss was about 5~10 %). The second activity test was carried out with the collected catalyst under the same reaction conditions as the first one (The catalyst amount was kept constant by decreasing the reaction numbers.)

## Characterization of CeO<sub>2</sub>, H-FAU, and CeO<sub>2</sub> + H-FAU

The specific surface area of the catalysts was measured by the Brunauer–Emmett–Teller (BET) isotherm analysis of N<sub>2</sub> physical adsorption using Gemini VII (Micromeritics). The X-ray powder diffraction patterns were obtained under air by miniflex600 (Rigaku) with Cu  $K_{\alpha}$  (40kV/15mA). The TEM images were recorded by the STEM/SEM HD-2700 (Hitachi). The preparation of TEM sample is that the catalyst was dispersed in ethanol under air, and then dropped on Cu microgrids.

	Reactant (	mmol)	Tommonotuno	CO <sub>2</sub>	Time	Viald <sup>a</sup>	
Catalyst	Methanol	DMP	(K)	Pressure (MPa)	(h)	(%)	Ref.
$Bu_2Sn(OMe)_2$	200	10	453	30	96	57	[S1]
$Bu_2Sn(OMe)_2$	200	10	453	200	24	88	[S1]
$Ce_{0.2}Zr_{0.8}O_2$	192	10	383	5	140	41	[S2]
Bu <sub>2</sub> Sn(OMe) <sub>2</sub> /[Ph <sub>2</sub> NH <sub>2</sub> ]-OTf	100	50	453	30	24	40	[S3]
Ti (OiPr) <sub>4</sub> /decyl-18- crown-6	200	10	473	30	24	58	[S4]
Spindle-CeO <sub>2</sub>	370	30	413	5	2	9	[S5]
$Ce_{0.5}Zr_{0.5}O_2$	618	10	413	7.5	3	236 <sup>b</sup>	[S6]

 Table S2 Previous reports of dimethyl carbonate (DMC) synthesis from CO2 and methanol with 2,2 

 dimethoxypropane (DMP) hydrolysis

<sup>a</sup>DMC yield based on DMP.

<sup>b</sup>The DMC yield based on DMP was higher than 100 % because the formed DMC amount was larger than that of the introduced DMP.

Table S3 Details of diethyl carbonate (DEC) synthesis from CO<sub>2</sub> and ethanol over CeO<sub>2</sub> with or without 2,2-diethoxypropane (DEP)

(a) DEC synthesis from  $CO_2$  and ethanol over  $CeO_2$  without DEP

Peaction time	Product /mmol	Ethanol balance
/h	DEC	/%
1	0.134	93
2	0.150	99
3	0.143	100
4	0.157	98
24	0.155	98

Reaction conditions: CeO<sub>2</sub> 0.5 g, ethanol 140 mmol, CO<sub>2</sub> 6.5 MPa (393 K), 393 K.

(b) DEC synthesis from  $CO_2$  and ethanol over  $CeO_2$  with DEP

Reaction time	DEP conversion	_		Products /mm	DEC Yield <sup>a</sup>	DEP balance		
/h	/%	DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	/%	/%
1	10	0.4	1.1	1.2	< 0.1	< 0.1	2	108
4	7	0.8	1.0	0.4	< 0.1	< 0.1	4	91
10	15	1.3	2.3	0.8	< 0.1	< 0.1	6	99
16	17	1.7	2.5	0.9	< 0.1	< 0.1	8	105
24	19	1.9	2.9	1.1	< 0.1	< 0.1	9	101
36	18	2.2	3.1	0.8	< 0.1	< 0.1	10	106

Reaction conditions: CeO<sub>2</sub> 0.5 g, ethanol 140 mmol, DEP 21 mmol, CO<sub>2</sub> 6.5 MPa (393 K), temperature 393 K.

<sup>a</sup>DEC yield based on DEP.

_			DFP conversion			Products	DFC Yield <sup>a</sup>	DEP balance		
Entry	Acid additive	pK <sub>a</sub>	/%	DEC	Acetone	2-Ethoxy propene	Mesityl oxide	Others <sup>b</sup>	/%	/%
1	-		16	0.8	1.0	0.4	< 0.1	< 0.1	4	91
2	HNO <sub>3</sub>	-1.3	25	2.3	3.2	0.4	< 0.1	< 0.1	11	92
3	$H_2SO_4$	-3.0, 1.9	56	9.9	10.0	0.3	< 0.1	< 0.1	48	93
4	H <sub>3</sub> PO <sub>4</sub>	2.1, 7.2, 12.3	20	1.1	1.8	0.5	< 0.1	< 0.1	5	91
5	Benzenesulfonic acid monohydrate	0.7	19	0.7	1.6	0.7	<0.1	< 0.1	3	92
6	Maleic acid	1.9, 6.2	21	0.6	2.1	0.7	< 0.1	< 0.1	3	92
7	Pyridine-2-carboxylic acid	3.1	18	0.4	1.6	0.7	< 0.1	< 0.1	2	93
8	Benzoic acid	4.2	20	0.6	1.9	0.5	< 0.1	< 0.1	3	91
9	H-FAU	-	65	13.1	13.4	0.2	<0.1	0.6	62	103

Table S4 Comparison of different homogeneous acid additives of CeO<sub>2</sub> in DEC synthesis with DEP hydrolysis

Reaction conditions: CeO<sub>2</sub> (873 K 3 h) 0.5 g, additive amount: 0.05 mmol (HNO<sub>3</sub>, benzenesulfonic acid monohydrate, pyridine-2-carboxylic acid, benzoic acid, *p*-toluenesulfonic acid) 0.025 mmol (H<sub>2</sub>SO<sub>4</sub>, H<sub>3</sub>PO<sub>4</sub>, maleic acid), 393 K, 4 h, CO<sub>2</sub> 6.5 MPa (393 K).

<sup>a</sup>DEC yield based on DEP.

Enter	H-FAU amount	DEP conversion			Products /n	DEC Yield <sup>a</sup>	DEP balance		
Entry	/g	/%	DEC A	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	/%	/%
1	0	10	0.4	1.1	1.2	< 0.1	< 0.1	2	108
2	0.002	30	3.6	4.1	0.5	< 0.1	< 0.1	17	91
3	0.005	44	6.4	6.7	0.4	< 0.1	0.2	30	91
4	0.010	42	8.2	8.6	0.4	< 0.1	< 0.1	40	101
5	0.020	65	10.9	10.8	0.2	< 0.1	0.7	51	92
6	0.050	74	11.8	11.7	0.1	0.1	1.2	56	89

Table S5 Effect of H-FAU amount in DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> + H-FAU

Reaction conditions: catalyst: CeO<sub>2</sub> (873 K 3 h) 0.5 g, CO<sub>2</sub> 6.5 MPa (393 K), 1 h.

<sup>a</sup>DEC yield based on DEP.

Enter	Reaction time	DEP conversion	_		Products /n	DEC yield <sup>a</sup>	DEP balance		
Entry	/h	/%	DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	/%	/%
1	0.5	37	6.6	6.6	0.6	< 0.1	< 0.1	31	97
2	1	42	8.2	8.6	0.4	< 0.1	< 0.1	40	101
3	4	65	13.1	13.4	0.2	< 0.1	0.6	62	103
4	24	67	13.0	13.1	0.1	<0.1	0.3	62	98

Table S6 Details of time-course of DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> + H-FAU (Figure 2)

Reaction conditions: CeO<sub>2</sub> 0.5 g, H-FAU 0.01 g, ethanol 140 mmol, DEP 21 mmol, 393 K. CO<sub>2</sub> 6.5 MPa (393 K).

<sup>a</sup>DEC yield based on DEP.

Entry Dausa tima		DEP conversion			Products /n	DEC yield <sup>a</sup>	DEP balance		
Епиу	Keuse unie	/%	DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	/%	/%
1	0	47	8.1	9.6	0.3	<0.1	0.1	39	101
2	1	45	8.2	9.1	0.4	< 0.1	0.1	39	101
3	2	45	8.3	9.2	0.4	< 0.1	0.1	39	101
4	3	40	8.2	9.2	0.4	< 0.1	< 0.1	39	106

Table S7 Recyclability test of CeO<sub>2</sub> + H-FAU in DEC synthesis with the presence of DEP (Figure 3)

Reaction conditions: CeO<sub>2</sub> 0.5 g, ethanol 140 mmol, DEP 21 mmol, H-FAU 0.01 g (reusing times: 0), mixed catalyst 0.5 g (reusing times 1~3), 393 K, CO<sub>2</sub> 6.5 MPa (393 K), 1 h.

<sup>a</sup>DEC yield based on DEP.



Figure S1 XRD patterns and specific surface area of fresh  $CeO_2$ , H-FAU and mixture of  $CeO_2$  and H-FAU before and after reaction

 $CeO_2$ : H-FAU(1) = 50 : 1 (weight ratio).

\*Specific surface areas were determined by BET method.



(a) H-FAU, (b) CeO<sub>2</sub>, (c) Mixture of CeO<sub>2</sub> and H-FAU after reaction, (d) Expanded figure of (c), (e) Mixture of CeO<sub>2</sub> and H-FAU after reaction (another view), (f) Expanded figure of (e) Reaction conditions: CeO<sub>2</sub> 0.5 g, ethanol 140 mmol, DEP 21 mmol, H-FAU 0.01 g, 393 K, CO<sub>2</sub> 6.5 MPa 393 K), 1 h.

Enters	EtOH amount				Products / mr	DEC vield <sup>a</sup> $/\%$	DED halanaa /0/			
Entry	/mmol	DEP conversion / %	DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	DEC yield"/%	DEI balance / 70	
$1^c$	0	16	0.3	1.3	1.6	0.3	0.3	1	86	
2	20	21	0.7	1.5	2.5	0.3	< 0.1	3	91	
3	60	34	4.4	4.6	0.9	0.2	0.3	21	96	
4	100	39	7.4	7.7	0.6	0.1	0.4	35	103	
5	140	42	8.2	8.6	0.4	< 0.1	< 0.1	40	101	
6	180	44	9.5	10.3	0.4	< 0.1	< 0.1	45	107	

Table S8 Effect of ethanol amount in DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> + H-FAU

Reaction conditions: CeO<sub>2</sub> 0.5 g, H-FAU 0.01 g, ethanol 0-180 mmol, DEP 21 mmol, 1 h, 393 K, CO<sub>2</sub> 6.5 MPa at 393 K.

<sup>a</sup>DEC yield based on DEP.

<sup>b</sup>Others include products derived from acetone.

<sup>c</sup>2.6 mmol ethanol was detected after the reaction

Entry Temp. /K	Т /И				Products /mm	DEC Vielda /0/	Balance /%		
	DEP COnversion /%	DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	DEC Yield <sup>a</sup> /%	DEP	
1	363	14	0.3	0.7	0.1	< 0.1	< 0.1	1	92
2	373	21	1.7	2.1	0.1	< 0.1	< 0.1	8	92
3	383	28	4.0	4.2	0.2	< 0.1	< 0.1	19	93
4	393	41	8.2	8.6	0.4	< 0.1	< 0.1	40	101
5	403	60	9.6	9.7	0.5	0.1	0.2	45	90

Table S9 Effect of reaction temperature in DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> and H-FAU

Reaction conditions: catalyst: CeO<sub>2</sub> 0.5 g, H-FAU 0.01 g, ethanol 140 mmol, DEP 21 mmol, CO<sub>2</sub> 6.5 MPa at 393 K, 1 h.

<sup>a</sup>DEC yield based on DEP.

Entry	CO <sub>2</sub> Pressure	Reaction time /h	DEP conversion /%			Products /mn	DEC vielda /%	DEP balance /%			
Linuy	at 393 K/MPa	Reaction time /ii		DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	DEC yield 7/0		
1	3.9	0.5	32	5.2	5.4	0.6	< 0.1	< 0.1	25	97	
2	3.9	1	36	7.2	7.3	0.3	< 0.1	< 0.1	34	101	
3	3.9	4	51	8.9	9.9	0.2	< 0.1	0.3	42	99	
4	3.9	24	54	10.4	10.6	0.2	< 0.1	< 0.1	49	98	
5	3.9	72	60	10.3	10.7	0.2	0.1	< 0.1	49	92	
6	6.5	0.5	37	6.6	6.6	0.6	< 0.1	< 0.1	31	97	
7	6.5	1	41	8.2	8.6	0.4	< 0.1	< 0.1	40	101	
8	6.5	4	65	13.1	13.4	0.2	< 0.1	0.6	62	103	
9	6.5	24	67	13.0	13.1	0.1	< 0.1	0.3	62	98	
10	6.5	72	69	12.7	12.3	0.2	0.1	< 0.1	60	91	
11	8.3	0.5	41	6.8	6.8	0.3	< 0.1	< 0.1	32	93	
12	8.3	1	40	8.3	8.6	0.5	< 0.1	< 0.1	41	103	
13	8.3	4	64	12.4	12.5	0.2	0.3	< 0.1	59	99	
14	8.3	24	77	13.1	13.2	0.2	0.2	0.7	63	92	
15	8.3	72	74	14.0	14.5	0.1	< 0.1	0.5	66	97	
16	9.5	0.5	32	6.6	6.7	0.7	< 0.1	< 0.1	29	103	
17	9.5	1	40	8.4	8.5	0.5	< 0.1	< 0.1	40	102	
18	9.5	4	60	11.3	11.4	0.2	< 0.1	0.1	53	96	
19	9.5	24	76	13.5	13.4	0.1	0.1	0.8	64	93	
20	9.5	72	74	15.2	15.3	0.2	0.1	0.7	72	104	

Table S10 Effect of CO<sub>2</sub> pressure on DEC synthesis from CO<sub>2</sub> and ethanol in the presence of DEP with CeO<sub>2</sub> + H-FAU (Figure 4)

Reaction conditions: catalyst: CeO<sub>2</sub> 0.5 g, H-FAU 0.01 g, ethanol 140 mmol, DEP 21 mmol, 393 K.

<sup>a</sup>DEC yield based on DEP.

Table S11 Details of comparison of DEC formation with different reaction systems in DEC synthesis

## from CO<sub>2</sub> and ethanol (Figure 5)

(a)	DEC for	nation from CO2 and C2H5OH with CeO2	2
		Draduata /mmal	

Destar	Time	Products /IIIIIoi				
Entry	/min	DEC				
1	0	0.006				
2	10	0.018				
3	20	0.031				

Reaction conditions: CeO<sub>2</sub> 0.03 g, ethanol 140 mmol, CO<sub>2</sub> 5 MPa (393 K).

Enter	Reaction time	Product /mmol			
Entry	/min	DEC			
1	0	0.004			
2	10	0.017			
3	20	0.030			

Reaction conditions: CeO<sub>2</sub> 0.03 g, ethanol 140 mmol, H-FAU 0.01 g, CO<sub>2</sub> 5 MPa (393 K).



(c) DEC formation from  $CO_2$  and  $C_2H_5OH$  in the presence of DEP with  $CeO_2$ 

<u> </u>	Reaction	DEP			Products /mmol		-	DEC	DEP	
Entry	time /min	conversion /%	DEC	Acetone	2-Ethoxypropene	Mesityl oxide	Others <sup>b</sup>	Yield <sup>a</sup> /%	balance /%	
1	0	3	0.007	0.1	0.4	< 0.1	< 0.1	<1	94	
2	7	4	0.021	0.2	0.6	< 0.1	< 0.1	<1	96	
3	17	5	0.044	0.2	0.8	< 0.1	< 0.1	<1	96	

Reaction conditions: CeO<sub>2</sub> 0.03 g, ethanol 140 mmol, DEP 21 mmol, CO<sub>2</sub> 5 MPa (393 K).

<sup>a</sup>DEC yield based on DEP.

<sup>b</sup>Others include products derived from acetone.

(d) DEC formation from  $CO_2$  and  $C_2H_5OH$  in the presence of DEP with  $CeO_2 + H$ -FAU

	Reaction	DEP	Products /mmol		DEC	DEP				
Entry time conversion /min /%		conversion /%	DEC Acetone 2-Ethoxypropene			Mesityl oxide	Others <sup>b</sup>	Yield <sup>a</sup> /%	balance /%	
1	0	9	0.096	0.8	0.8	< 0.1	< 0.1	<1	97	
2	7	10	0.278	1.1	0.8	< 0.1	< 0.1	1	101	
3	17	10	0.470	1.3	0.6	< 0.1	< 0.1	2	100	

Reaction conditions: CeO<sub>2</sub> 0.03 g, H-FAU 0.01 g, ethanol 140 mmol, DEP 21 mmol, CO<sub>2</sub> 5 MPa (393 K). <sup>*a*</sup>DEC yield based on DEP.





Figure S3(c) The DEC formation rate of  $CO_2 + C_2H_5OH + DEP$  on  $CeO_2$ 

Figure S3(d) The DEC formation rate of  $CO_2 + C_2H_5OH + DEP$  on  $CeO_2 + H$ -FAU



Figure S4 (a) Typical GC chart of DEC synthesis from  $CO_2$  and ethanol in the presence of DEP with  $CeO_2$  and H-FAU

Reaction conditions: CeO<sub>2</sub> 0.5 g, H-FAU 0.01 g, ethanol 140 mmol, DEP 21 mmol, 393 K, 1 h, CO<sub>2</sub> 6.5 MPa (393 K)

Retention time (min)	Compound
2.946	2-Ethoxypropene
3.456	Acetone
4.334	2,2-Diethoxypropane
5.459	Ethanol
8.577	Diethyl carbonate
8.935	Mesityl oxide
10.129, 10.637	Polymerized products of acetone
11.327	1-Hexanol (internal standard)



Figure S4 (b) GC chart of entry 20 in Table S10

Reaction conditions: CeO<sub>2</sub> 0.5 g, H-FAU 0.01 g, ethanol 140 mmol, DEP 21 mmol, 393 K, 72 h, CO<sub>2</sub> 9.5 MPa (393 K)

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