**Electronic Supporting Information (ESI):**

**Fatty acid based bicarbonates: Al-mediated stereoselective preparation of mono-, di- and tricarbonates under mild and solvent-less conditions**

L. Peña Carrodeguas, a À. Cristòfol, a J. M. Fraile, b J. A. Mayoral, b V. Dorado, b C. I. Herrerías b, * and A. W. Kleij a, c, *

a Institute of Chemical Research of Catalonia (ICIQ), the Barcelona Institute of Science and Technology, Av. Països Catalans 16, 43007 – Tarragona (Spain). E-mail: akleij@iciq.es

b Departamento de Catálisis y Procesos Catalíticos, Instituto de Síntesis Química y Catálisis Homogénea (ISQCH), CSIC-Universidad de Zaragoza, Facultad de Ciencias, C/ Pedro Cerbuna 12, 50009 Zaragoza, Spain. E-mail: clarah@unizar.es

c Catalan Institute for Research and Advanced Studies (ICREA), Pg. Lluis Companys 23, 08010 – Barcelona (Spain).

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1. General Considerations:

The Al(III) aminotriphenolate complexes and the starting ligands were synthesized according to reported literature procedures.\textsuperscript{[1,2]} Commercially available fatty acids, solvents, co-catalysts, oxidants were purchased from various commercial sources (Acros, Aldrich and TCI) and used without further purification. Carbon dioxide (purchased from PRAXAIR) was used without further purification or drying prior to its use.

\textsuperscript{1}H NMR and \textsuperscript{13}C NMR spectra were recorded at rt on a Bruker AV-300, AV-400 or AV-500 spectrometer and referenced to the residual deuterated solvent signals. Diastereo-isomeric ratios (dr’s) were calculated from the corresponding \textsuperscript{1}H NMR spectra using signal integration where possible; alternatively, integrable \textsuperscript{13}C NMR spectra were recorded with a 500 MHz AV-500 spectrometer. All reported NMR values are given in parts per million (ppm). FT-IR measurements were carried out on a Bruker Optics FTIR Alpha spectrometer equipped with a DTGS detector, KBr beam splitter at 4 cm\textsuperscript{-1} resolution. Mass spectrometric analyses were performed by the Research Support Group at ICIQ.

2. Experimental Procedures:

Acid protection method (typical procedure):

1 g of fatty acid and 5 mg of para-toluenesulfonic acid (mono-hydrate) were dissolved in MeOH (50 mL). The reaction mixture was heated to reflux for 3 h. Hereafter, the solvent was removed in vacuo (rotary evaporator) and the fatty ester product was used without further purification in the next step – i.e. epoxidation.
Epoxidation method (typical procedure):

1 g of the methyl ester was dissolved in CH₂Cl₂ (40 mL) and cooled to 0 °C. Then meta-chloroperbenzoic acid (1.2 equiv) was slowly added to this solution. The reaction mixture was left stirring for 12 h at rt. The solution was then washed with an aqueous solution of Na₂SO₃ (3 × 30 mL, 1 M), a saturated aqueous solution of NaHCO₃ (3 × 30 mL) and brine. The organic phase was dried over sodium sulfate and the product was obtained after removal of the solvent in vacuo using a rotary evaporator.

Carbonate formation (Screening experiments):
The screening experiments of all fatty acid carbonate syntheses were done by using a HEL pressure multi-reactor (HEL CAT²⁴) with 24 vials of 10 × 75 mm. The epoxide substrate (0.32 mmol) was introduced into a vial of the HEL with a pre-selected loading of catalyst and co-catalyst and solvent volume. The HEL was pressurized to 10 bar with CO₂ and heated to the desired temperature. The reaction mixture was stirring for 24-48 h. After cooling to ambient temperature, the crude product was analyzed by ¹H NMR (CDCl₃) to determine the conversion, selectivity and cis/trans ratio.

Carbonate formation (scaled-up experiments to isolate):
The epoxide substrate (0.5 g) was introduced into a 30 mL autoclave with a pre-selected loading of catalyst (Al-complex) and co-catalyst (nucleophile) and solvent where required. The autoclave was heated to the desired temperature, pressurized to 10 bar with CO₂ and the reaction mixture left stirring for 24–48 h. Then, after cooling to ambient temperature, the reaction was carefully and slowly vented, and the crude product analyzed by ¹H NMR and ¹³C NMR (CDCl₃). The product was purified by flash chromatography (silica gel, EtOAc/hexane) following subsequent removal of the solvent using rotary evaporation.
3. $^1$H-NMR and $^{13}$C-NMR Data of Epoxide Products:

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$: 3.66 (s, 3H), 2.89 (m, 2H), 2.30 (t, $^3$$J_{H,H}$ = 7.5 Hz, 2H), 1.67-1.56 (m, 2H), 1.53-1.20 (m, 24H), 0.87 (t, $^3$$J_{H,H}$ = 6.7 Hz, 3H).

$^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$: 174.24 (C=O), 57.25 (CH), 57.20 (CH), 51.46 (OCH$_3$), 34.11 (CH$_2$), 31.92 (CH$_2$), 29.62 (CH$_2$), 29.60 (CH$_2$), 29.40 (CH$_2$), 29.29 (CH$_2$), 29.24 (CH$_2$), 29.10 (CH$_2$), 27.91 (CH$_2$), 27.87 (CH$_2$), 26.68 (CH$_2$), 26.63 (CH$_2$), 24.96 (CH$_2$), 22.73 (CH$_2$), 14.14 (CH$_3$).

MS (ESI+, CH$_3$OH): $m/z$ calcd. 335.3 (M+Na)$^+$, found: 335.3. [1]
Methyl 8-((3-(pentyloxiran-2-yl)methyl)oxiran-2-yl)octanoate (B in the main text). Note: mixture of two diastereoisomers.

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 3.65 (s, 3H), 3.12-3.02 (m, 2H), 2.99-2.92 (m, 2H), 2.28 (t, $^3$J$_{HH}$ = 7.5 Hz, 2H), 1.82-1.68 (m, 2H), 1.65-1.56 (m, 2H), 1.56-1.41 (m, 8H), 1.38-1.26 (m, 10H), 0.88 (t, $^3$J$_{HH}$ = 6.9 Hz, 3H)

$^{13}$C NMR (101 MHz, CDCl$_3$) $\delta$ 174.32 (C=O), 57.12 (CH), 57.07 (CH), 56.84 (CH), 56.78 (CH), 54.47 (CH), 54.45 (CH), 54.29 (CH), 51.55 (OCH$_3$), 34.16 (CH$_2$), 31.78 (CH$_2$), 29.40 (CH$_2$), 29.26 (CH$_2$), 29.13 (CH$_2$), 28.01 (CH$_2$), 28.00 (CH$_2$), 27.93 (CH$_2$), 27.92 (CH$_2$), 27.33 (CH$_2$), 27.05 (CH$_2$), 26.64 (CH$_2$), 26.53 (CH$_2$), 26.36 (CH$_2$), 26.25 (CH$_2$), 25.00 (CH$_2$), 22.67 (CH$_2$), 14.09 (CH$_3$).

MS (ESI+, CH$_3$OH): $m/z$ calcd. 349.2 (M+Na)$^+$, found: 349.2. $^{[3]}$
Methyl 8-((3-((3-ethylloxiran-2-yl)methyl)oxiran-2-yl)methyl)oxiran-2-yl)octanoate (C in the main text). Note: mixture of four diastereoisomers.

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 3.60 (s, 3H), 3.16-3.01 (m, 4H), 2.96-2.86 (m, 2H), 2.24 (t, $^3$J$_{H,H}$ = 7.3 Hz, 2H), 1.82-1.61 (m, 4H), 1.60-1.35 (m, 8H), 1.34-1.22 (m, 6H), 1.04-0.97 (m, 3H).

$^{13}$C NMR (101 MHz, CDCl$_3$) $\delta$ 174.19 (C=O), 58.14 (CH), 58.11 (CH), 57.83 (CH), 56.95 (CH), 56.93 (CH), 56.64 (CH), 54.26 (CH), 54.18 (CH), 54.11 (CH), 54.05 (CH), 53.99 (CH), 53.78 (CH), 51.42 (OCH$_3$), 34.02 (CH$_2$), 29.26 (CH$_2$), 29.13 (CH$_2$), 28.99 (CH$_2$), 27.88 (CH$_2$), 27.82 (CH$_2$), 27.36 (CH$_2$), 27.29 (CH$_3$), 27.24 (CH$_3$), 27.16 (CH$_3$), 27.03 (CH$_3$), 26.97 (CH$_3$), 26.92 (CH$_3$), 26.85 (CH$_3$), 26.51 (CH$_3$), 26.42 (CH$_3$), 26.40 (CH$_3$), 24.87 (CH$_2$), 21.25 (CH$_3$), 21.22 (CH$_3$), 21.14 (CH$_2$), 10.59 (CH$_3$), 10.48 (CH$_3$).

HRMS (ESI+, CH$_3$OH): m/z calcd. 363.2142 (M+Na)$^+$, found: 363.2147.
Methyl 12-3-octyloxiran-2-yl)dodecanoate (D in the main text).

$^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 3.66 (s, 3H), 2.92-2.87 (s, 2H), 2.30 (t, $^3J_{H,H}=7.5$ Hz, 2H), 1.65-1.59 (m, 2H), 1.53-1.45 (m, 6H), 1.39-1.19 (m, 26H), 0.88 (t, $^3J_{H,H}=7.1$ Hz, 3H).

$^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 174.46 (C=O), 57.39 (CH), 51.57 (OCH$_3$), 34.26 (CH$_2$), 31.01 (CH$_2$), 29.70 (CH$_2$), 29.68 (CH$_2$), 29.67 (CH$_2$), 29.58 (CH$_2$), 29.40 (CH$_2$), 29.37 (CH$_2$), 29.30 (CH$_2$), 29.98 (CH$_2$), 27.06 (CH$_2$), 26.75 (CH$_3$), 25.11 (CH$_3$), 22.81 (CH$_2$), 14.24 (CH$_3$).

MS (ESI+, CH$_3$OH): $m/z$ calcd. 391.3 (M+Na)$^+$, found: 391.3. $^{[3]}$
Methyl 8-3-octyloxiran-2-yl)octanoate (E in the main text).

^1H NMR (500 MHz, CDCl$_3$) $\delta$ 3.59 (s, 3H), 2.57 (m, 2H), 2.23 (t, $^3J_{HH} = 7.4$ Hz, 2H), 1.58-1.51 (m, 2H), 1.47-1.40 (m, 4H), 1.40-1.31 (m, 4H), 1.30-1.16 (m, 16H), 0.81 (t, $^3J_{HH} = 6.9$ Hz, 3H).

^13C NMR (101 MHz, CDCl$_3$) $\delta$ 174.10 (C=O), 58.80 (CH), 58.75 (CH), 51.34 (OCH$_3$), 34.01 (CH$_2$), 32.13 (CH$_2$), 32.09 (CH$_2$), 31.85 (CH$_2$), 29.52 (CH$_2$), 29.46 (CH$_2$), 29.23 (CH$_2$), 29.16 (CH$_2$), 29.02 (CH$_2$), 26.06 (CH$_2$), 25.99 (CH$_2$), 24.88 (CH$_2$), 22.65 (CH$_2$), 14.07 (CH$_3$).

HRMS (ESI+, CH$_3$OH): m/z calcd. 251.0526 (M+Na)$^+$, found: 251.0527.
Methyl 8-(3-(2-hydroxyoctyl)oxiran-2-yl)octanoate (F in the main text). Note: mixture of two diastereoisomers ($dr = 56:44$).

$^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 3.86-3.74 (m, 1H), 3.61 (s, 3H), 3.11-3.05 (m, 1H), 2.93-2.82 (m, 1H), 2.60-2.35 (2 x br s, 1H, OH), 2.25 (t, $^3$J$_{H,H} = 7.5$ Hz, 2H), 1.77-1.64 (m, 1H), 1.61-1.54 (m, 2H), 1.51-1.42 (m, 6H), 1.33-1.20 (m, 15H), 0.83 (t, $^3$J$_{H,H} = 6.8$ Hz, 3H)

$^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 174.19 (C=O), 174.18 (C=O), 70.65 (CH), 69.87 (CH), 57.12 (CH), 56.31 (CH), 55.25 (CH), 54.50 (CH), 51.38 (OCH$_3$), 37.79 (CH$_2$), 37.42 (CH$_2$), 35.25 (CH$_2$), 34.84 (CH$_2$), 34.00 (CH$_2$), 31.80 (CH$_2$), 29.28 (CH$_2$), 29.27 (CH$_2$), 29.25 (CH$_2$), 29.23 (CH$_2$), 29.12 (CH$_2$), 28.97 (CH$_2$), 28.00 (CH$_2$), 27.87 (CH$_2$), 26.41 (CH$_2$), 26.38 (CH$_2$), 25.58 (CH$_2$), 25.51 (CH$_2$), 24.85 (CH$_2$), 22.58 (CH$_2$), 14.04 (CH$_3$).

MS (ESI+, CH$_3$OH): m/z calcd. 351.3 (M+Na)$^+$, found: 351.3. $^{[3]}$
4. \(^1\)H-NMR and \(^{13}\)C-NMR Data of Carbonate Products and MS Analysis.

Methyl 8-(5-octyl-2-oxo-1,3-dioxolan-4-yl)octanoate (5 in the main text).

For the major isomer of 5 (cis): \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 4.55-4.50 (m, 2H), 3.53 (s, 3H), 2.18 (t, \(^3\)J\(_{HH}\)= 7.5 Hz, 2H), 1.60-1.40 (m, 8H), 1.30-1.10 (m, 18H), 0.76 (t, \(^3\)J\(_{HH}\)= 7.1 Hz, 3H). The peak at \(\delta\) 4.14-4.11 (m) corresponds to trans-5 (3%).

For the major isomer of 5 (cis): \(^{13}\)C NMR (126 MHz, CDCl\(_3\)) \(\delta\) 173.80 (C=O), 154.51 (C=O), 79.78 (CH), 79.75 (CH), 51.13 (OCH\(_3\)), 33.74 (CH\(_2\)), 31.61 (CH\(_2\)), 29.15 (CH\(_2\)), 29.05 (CH\(_2\)), 28.96 (CH\(_2\)), 28.82 (CH\(_2\)), 28.79 (CH\(_2\)), 28.73 (CH\(_2\)), 28.68 (CH\(_2\)), 28.64 (CH\(_2\)), 25.44 (CH\(_2\)), 25.37 (CH\(_2\)), 24.63 (CH\(_2\)), 22.43 (CH\(_2\)), 13.86 (CH\(_3\)).

MS (ESI+, CH\(_3\)OH): \(m/z\) calcd. 379.2 (M+Na\(^+\), found: 379.2. \[^3\]
Methyl 8-(2-oxo-5-((2-oxo-5-pentyl-1,3-dioxolan-4-yl)methyl)-1,3-dioxolan-4-yl)octanoate (6 in the main text). Note: mixture of diastereoisomers, two *cis* diastereoisomers (97%) and two *trans* diastereoisomers (3%).

For the major isomers of 6 (*cis*): ^1^H NMR (400 MHz, CDCl$_3$) $\delta$ 4.90-4.65 (m, 4H), 3.60 (s, 3H), 2.24 (t, $^3$J$_{HH}$ = 7.6 Hz, 2H), 1.88-1.81 (m, 2H), 1.67-1.48 (m, 8H), 1.37-1.22 (m, 12H), 0.84 (t, $^3$J$_{HH}$ = 7.2 Hz, 3H). The peaks at $\delta$ 4.47-4.25 (m) correspond to *trans*-6 (3%).

For the major isomers of 6 (*cis*): ^1^C NMR (126 MHz, CDCl$_3$) $\delta$ 174.09 (C=O), 153.87 (C=O), 153.85 (C=O), 79.55 (CH), 79.50 (CH), 75.53 (CH), 51.39 (OCH$_3$), 33.91 (CH$_2$), 31.21 (CH$_2$), 28.89 (CH$_2$), 28.85 (CH$_2$), 28.77 (CH$_2$), 25.56 (CH$_2$), 25.30 (CH$_2$), 24.75 (CH$_2$), 22.33(CH$_2$), 13.85 (CH$_3$).

MS (ESI+, CH$_3$OH): m/z calcd. 437.2 (M+Na)$^+$, found: 437.2. $^{[3]}$
Methyl 8-((5-((5-ethyl-2-oxo-1,3-dioxolan-4-yl)methyl)-2-oxo-1,3-dioxolan-4-yl)methyl)-2-oxo-1,3-dioxolan-4-yl)octanoate (7). Note: mixture of diastereoisomers, four cis diastereoisomers (96%) and four trans diastereoisomers (4%).

For the major isomers of 7 (cis): $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 5.06-4.66 (m, 6H), 3.65 (s, 3H), 2.29 (t, $^3$J$_{H,H}$ = 7.4 Hz, 2H), 2.23-1.93 (m, 4H), 1.80-1.50 (m, 6H), 1.40-1.27 (m, 8H), 1.09 84 (t, $^3$J$_{H,H}$ = 7.2 Hz, 3H). The peaks at $\delta$ 4.55-4.15 (m) correspond to trans-7 (4%).

For the major isomers of 7 (cis): $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 174.33 (C=O), 154.19 (C=O), 154.11 (C=O), 153.44 (C=O), 81.24 (CH), 81.10 (CH), 79.90 (CH), 79.77 (CH), 76.63 (CH), 75.68 (CH), 51.59 (OCH$_3$), 34.08 (CH$_2$), 29.03 (CH$_3$), 29.03 (CH$_2$), 28.95 (CH$_3$), 28.88 (CH$_2$), 28.53 (CH$_3$), 28.25 (CH$_2$), 25.79 (CH$_3$), 25.62 (CH$_3$), 24.89 (CH$_2$), 24.89 (CH$_3$), 22.47 (CH$_2$), 22.40 (CH$_3$), 10.33 (CH$_3$).
HRMS (ESI+, CH$_3$OH): m/z calcd. 495.1837 (M+Na)$^+$, found: 495.1848.

Methyl 12-(5-octyl-2-oxo-1,3-dioxolan-4-yl)dodecanoate (8).

For the major isomer of 8 (cis): $^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 4.59-4.53 (m, 2H), 3.57 (s, 3H), 2.22 (t, $^3$$J_{H,H}$ = 7.5 Hz, 2H), 1.65-1.40 (m, 8H), 1.33-1.14 (m, 26H), 0.80 (t, $^3$$J_{H,H}$ = 7.1 Hz, 3H). The peak at $\delta$ 4.17-4.14 (m) corresponds to trans-8 (4%).

For the major isomer of 8 (cis): $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 174.07 (C=O), 154.66 (C=O), 79.90 (CH), 51.25 (OCH$_3$), 33.96 (CH$_2$), 31.72 (CH$_3$), 29.40 (CH$_2$), 29.36 (CH$_2$), 29.31 (CH$_2$), 29.27 (CH$_2$), 29.26 (CH$_2$), 29.17 (CH$_3$), 29.15 (CH$_3$), 29.07 (CH$_2$), 29.03 (CH$_2$), 28.80 (CH$_2$), 25.54 (CH$_2$), 24.85 (CH$_2$), 22.55 (CH$_2$), 13.98 (CH$_3$).

MS (ESI+, CH$_3$OH): m/z calcd. 435.3 (M+Na)$^+$, found: 435.3. [3]
Methyl 8-5-octyl-2-oxo-1,3-dioxolan-4-yl)octanoate (9) – dr >99:1.

$^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 4.22-4.19 (m, 2H), 3.66 (s, 3H), 2.29 (t, $^3$J$_{H,H}$ = 7.6 Hz, 2H), 1.76-1.67 (m, 2H), 1.66-1.56 (m, 4H), 1.52-1.41 (m, 2H), 1.35-1.24 (m, 18H), 0.87 (t, $^3$J$_{H,H}$ = 7.0 Hz, 3H).

$^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 174.29 (C=O), 154.82 (C=O), 82.14 (CH), 82.10 (CH), 51.58 (OCH$_3$), 34.11 (CH$_2$), 33.94 (CH$_2$), 33.93 (CH$_2$), 31.90 (CH$_2$), 29.42 (CH$_2$), 29.29 (CH$_2$), 29.23 (CH$_2$), 29.08 (CH$_2$), 29.03 (CH$_2$), 24.93 (CH$_2$), 24.76 (CH$_2$), 24.72 (CH$_2$), 22.74 (CH$_2$), 14.19 (CH$_3$).

HRMS (ESI+, CH$_3$OH): m/z calcd. 379.2455 (M+Na)$^+$, found: 379.2458.
Methyl 8-((5-(2-hydroxyoctyl)-2-oxo-1,3-dioxolan-4-yl)octanoate (10). Note: mixture of four diastereoisomers.

For the major isomers of 10 (trans): $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 4.55-4.23 (m, 2H), 3.81-3.69 (m, 1H), 3.58 (s, 3H), 2.62-2.49 (2 × br s, 1H, OH), 2.23 (t, $^3J_{H,H} = 7.4$ Hz, 2H), 1.91-1.19 (m, 24H), 0.81 (t, $^3J_{H,H} = 7.1$ Hz, 3H).
For the major isomers of 10 (trans): $^{13}$C NMR (126 MHz, CDCl$_3$) δ 174.25 (C=O), 154.80 (C=O), 154.77 (C=O), 82.52 (CH), 82.10 (CH), 79.84 (CH), 79.64 (CH), 67.80 (CH), 67.61 (CH), 51.40(OCH$_3$), 41.29 (CH$_2$), 40.19 (CH$_2$), 38.10 (CH$_2$), 37.49 (CH$_2$), 33.91 (CH$_2$), 33.50 (CH$_2$), 33.25 (CH$_2$), 31.70 (CH$_2$), 29.14 (CH$_2$), 28.84 (CH$_2$), 28.80 (CH$_2$), 25.36 (CH$_2$), 25.27 (CH$_2$), 24.74 (CH$_2$), 24.48 (CH$_2$), 22.51 (CH$_2$), 13.99 (CH$_3$).

MS (ESI+, CH$_3$OH): $m/z$ calcd. 395.2 (M+Na)$^+$, found: 395.2. [3]
5. $^1$H-NMR Analysis of the Crude Reaction Mixture for (5).

Figure S1. $^1$H-NMR reaction crude of cyclic carbonate 5 (data for the product obtained using the conditions reported in Table 1, entry 18). The NMR analysis shows the high chemo-selectivity towards cyclic carbonate formation and the virtual absence of methyl 9-oxooctadecanoate (5b) and methyl 10-oxooctadecanoate (5c), see below for a visual:

![Methyl 9-oxooctadecanoate (5b)](image)

![Methyl 10-oxooctadecanoate (5c)](image)

Figure S2. Methyl 9-oxooctadecanoate (5b) and Methyl 10-oxooctadecanoate (5c). Original data ($^1$H-NMR and $^{13}$C-NMR data) for 5b and 5c can be found in the Supplementary Information of reference [3].
6. IR Data of All Carbonate Products.

IR spectrum (neat) for 5.

IR spectrum (neat) for 6.
IR spectrum (neat) for 7.

IR spectrum (neat) for 8.
IR spectrum (neat) for 9.

IR spectrum (neat) for 10.
7. Kinetic Studies with and without Al Catalyst (3) for the Conversion of Oleate (A).

Figure S3:*

In blue: conversion of A into 5 in time using 3.2 mmol of A 0.5 mol% Al-complex 3, 3 mol% PPNCl, under neat conditions at 70°C and 10 bar initial CO₂ pressure.

In red: conversion of A into 5 in time under the same conditions in the absence of Al-complex 3.

*Please note that the different kinetic regimes are ascribed to a continuously changing reaction mixture (cf., polarity and viscosity) that may affect the CO₂ dissolution in the liquid phase and the kinetic behavior. Both profiles show the same tendency, but essentially one can notice a clear positive effect of the presence of the Al-complex throughout the reaction.
8. References:

