Supplementary material

Figure S1 Thermal diagrams (TGA/DSC) of starting and Al/Fe-pillared clays.

Figure S2 SEM micrographs (x500) for: (a) r; (b) r-LD; (c) R; (d) R-LC; (e) R-BC; (f) RI and (g) RI-PC clay catalysts; purple-squares highlight surface zones where Fe concentration was determined by EDS for variance analysis.

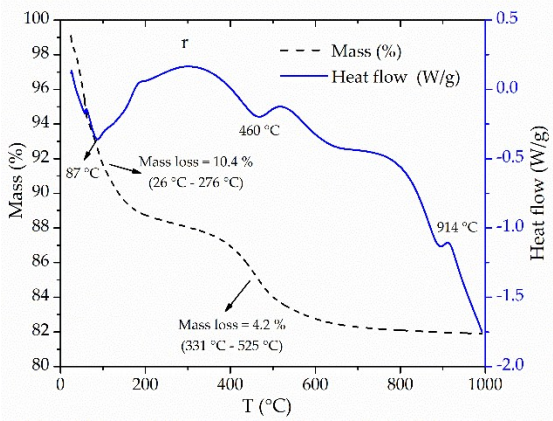
Figure S3 SEM micrographs (x2000) for: (a) r; (b) r-LD; (c) R; (d) R-LC; (e) R-BC; (f) RI and (g) RI-PC clay catalysts; purple-squares highlight surface zones where Fe concentration was determined by EDS for variance analysis.

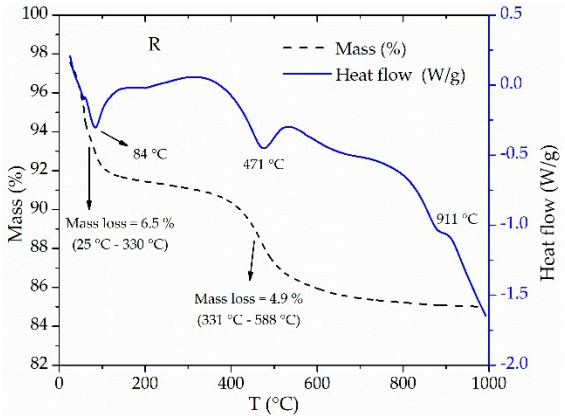
Figure S4 SEM micrographs (x4000) for: (a) r; (b) r-LD; (c) R; (d) R-LC; (e) R-BC; (f) RI and (g) RI-PC clay catalysts; purple-squares highlight surface zones where Fe concentration was determined by EDS for variance analysis.

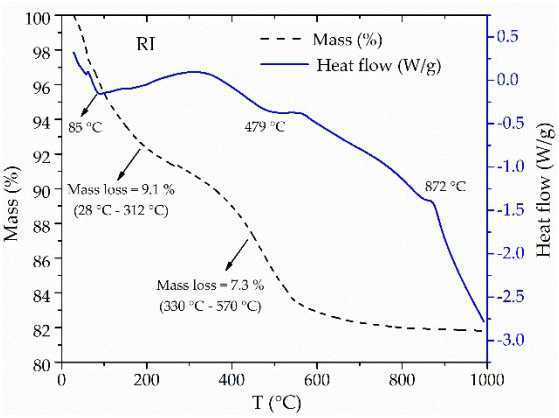
Figure S5 LC-DAD chromatograms of the (a) r-LD and (b) R-LC catalysts after 180 min of reaction (plus 30 min of pre-equilibrium period): comparison of diluted vs. concentrated precursors with oxalic acid detected.

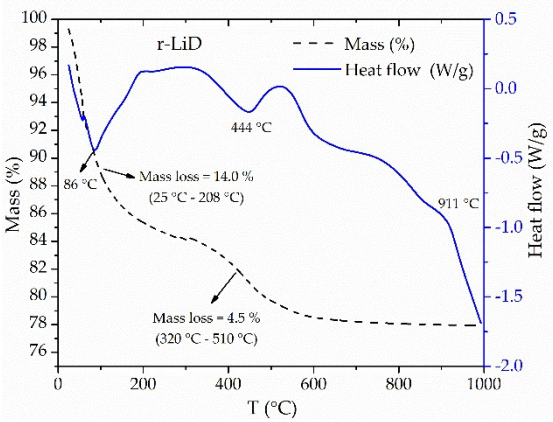
Figure S6 LC-DAD chromatograms of the (a) r-LD and (b) R-LC catalysts after 180 min of reaction (plus 30 min of pre-equilibrium period): comparison of diluted vs. concentrated precursors showing hydroquinone and *p*-benzoquinone expected retention times.

Figure S7 LC-DAD chromatograms of the (a) R-LC, (b) R-BC and (c) RI-PC catalysts after 180 min of reaction (plus 30 min of pre-equilibrium period): comparison lab- vs. bench- vs. pilot-scale preparations of the Al/Fe-PILC showing hydroquinone and *p*-benzoquinone expected retention times.


r
Mass (%)
Heat flow (W/g)
87 °C
Mass loss = 10.4 %
(26 °C - 276 °C)
460 °C
914 °C
Mass loss = 4.2 %
(331 °C - 525 °C)
Mass (%)
Heat flow (W/g)
T (°C)


R
Mass (%)
Heat flow (W/g)
84 °C
Mass loss = 6.5 %
(25 °C - 330 °C)
471 °C
911 °C
Mass loss = 4.9 %
(331 °C - 588 °C)
Mass (%)
Heat flow (W/g)
T (°C)


RI
Mass (%)
Heat flow (W/g)
85 °C
Mass loss = 9.1 %
(28 °C - 312 °C)
479 °C
872 °C
Mass loss = 7.3 %
(330 °C - 570 °C)
Mass (%)
Heat flow (W/g)
T (°C)


r-LiD
Mass (%)
Heat flow (W/g)
86 °C
Mass loss = 14.0 %
(25 °C - 208 °C)
444 °C
911 °C
Mass loss = 4.5 %
(320 °C - 510 °C)
Mass (%)
Heat flow (W/g)
T (°C)

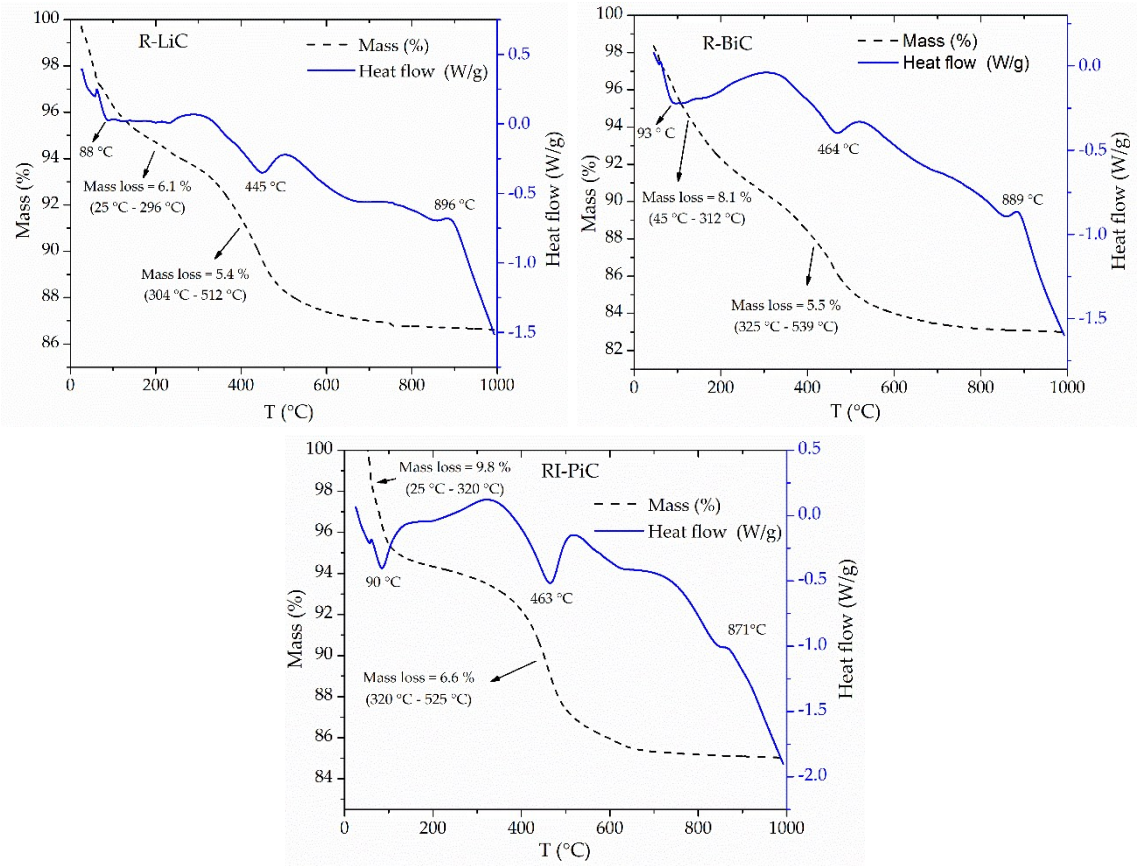
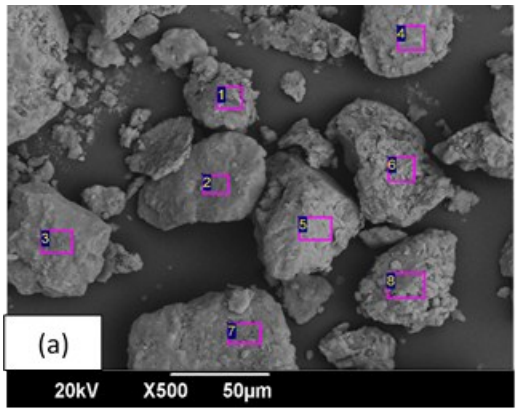
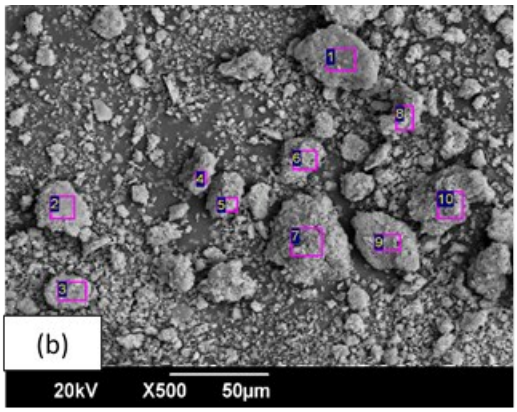
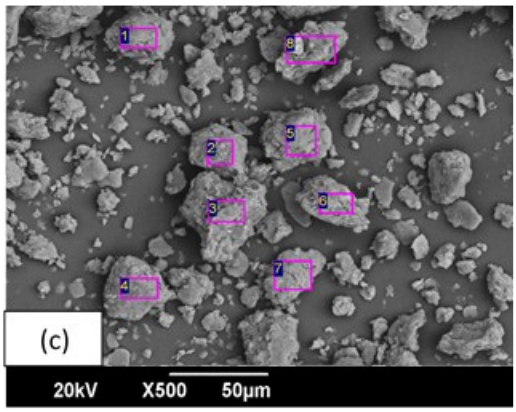
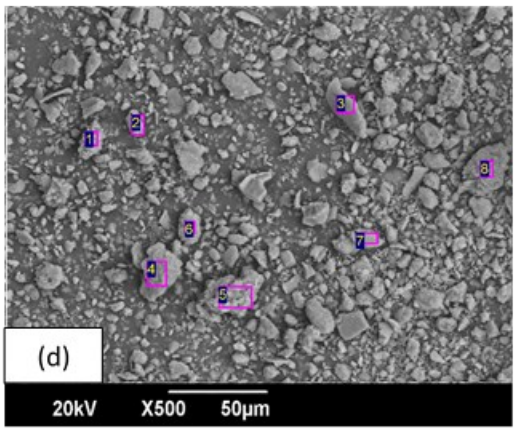


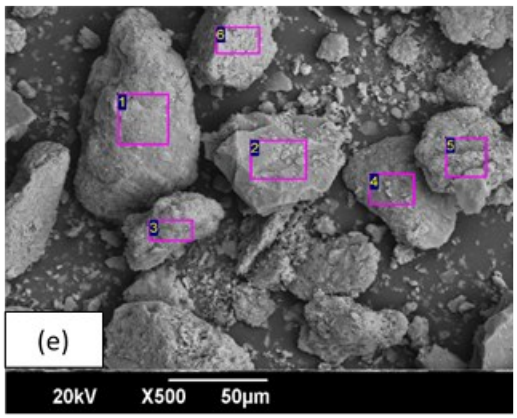
Figure S1 Thermal diagrams (TGA/DSC) of starting and Al/Fe-pillared clays.


(a)
20kV X500 50µm


(b)
20kV X500 50µm


(c)
20kV X500 50µm


(d)
20kV X500 50µm


(e)
20kV X500 50µm

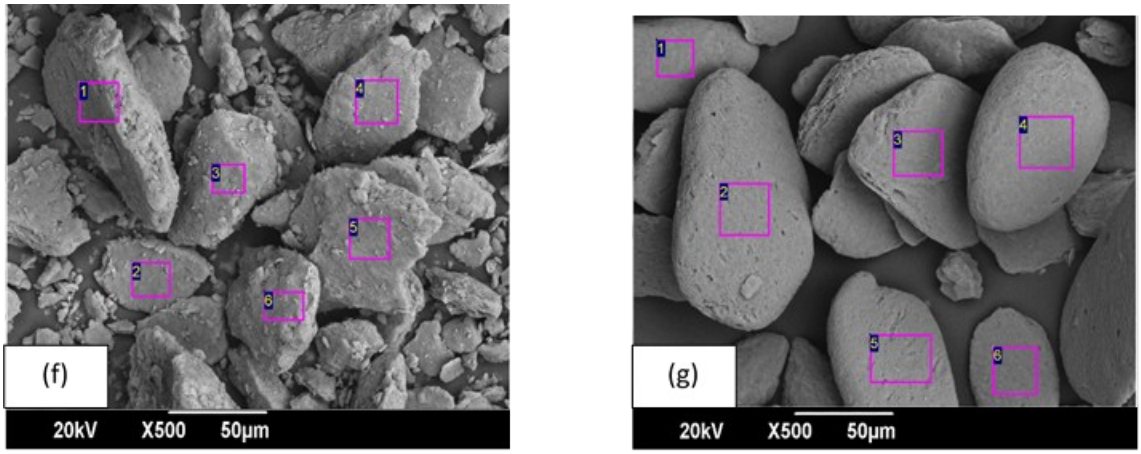
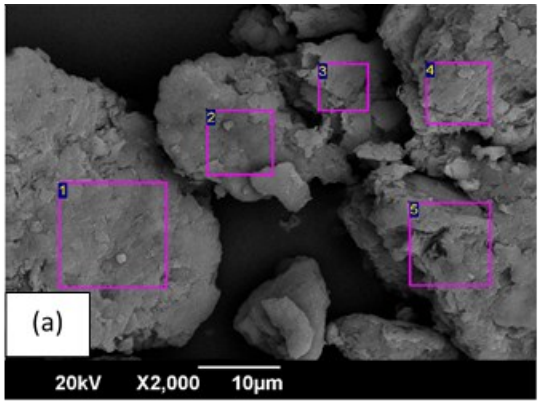
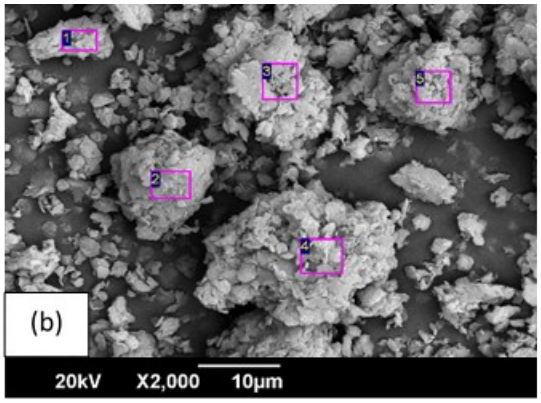
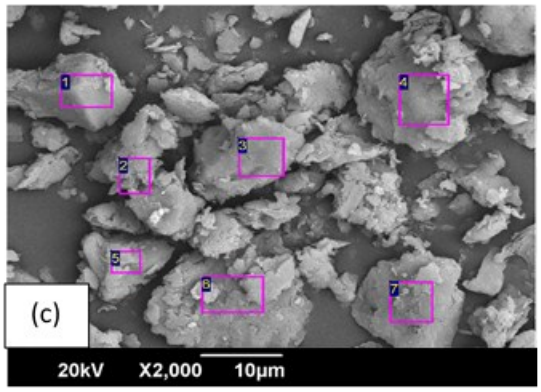
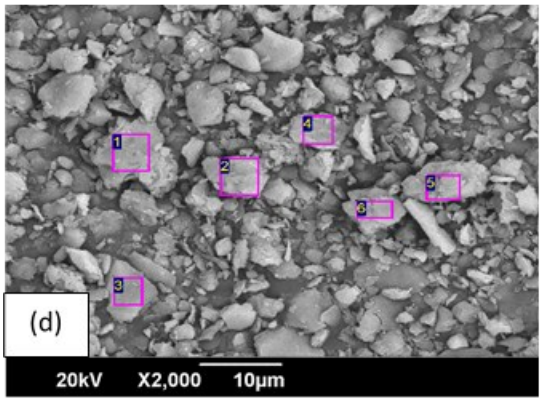


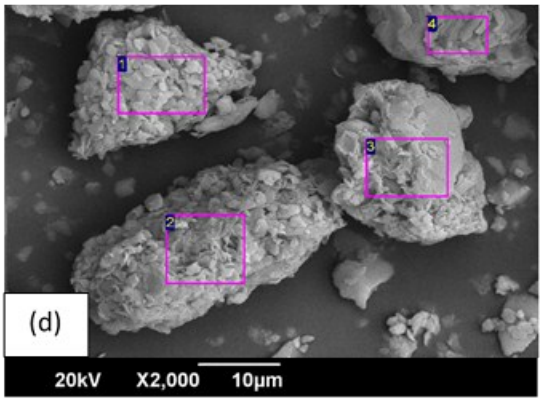
Figure S2 SEM micrographs (x500) for: (a) r; (b) r-LD; (c) R; (d) R-LC; (e) R-BC; (f) RI and (g) RI-PC clay catalysts; purple-squares highlight surface zones where Fe concentration was determined by EDS for variance analysis.


(a)
20kV X2,000 10µm


(b)
20kV X2,000 10µm


(c)
20kV X2,000 10µm


(d)
20kV X2,000 10µm


(d)
20kV X2,000 10µm

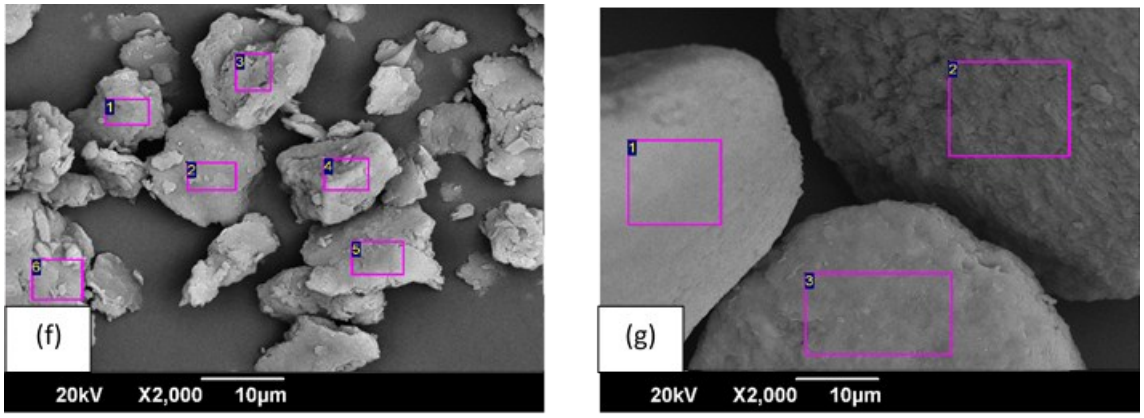
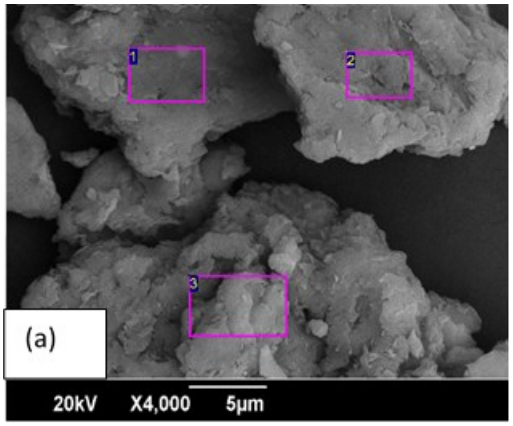
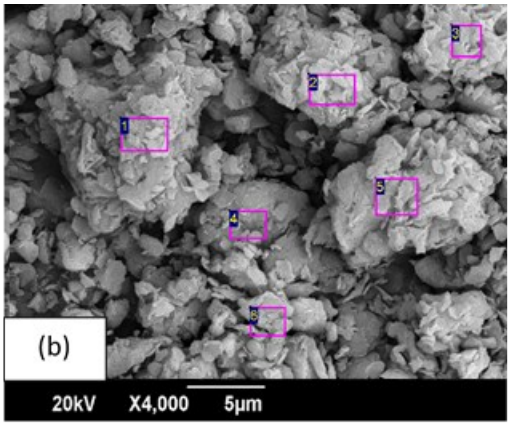
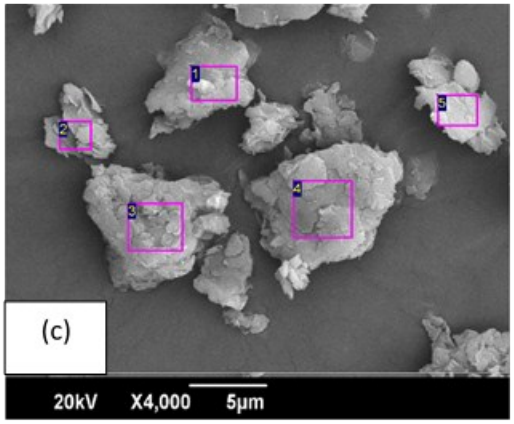
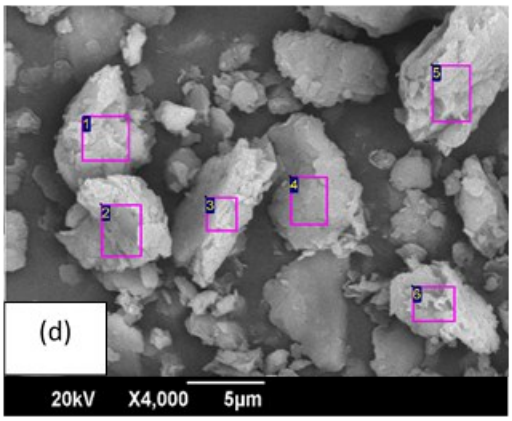


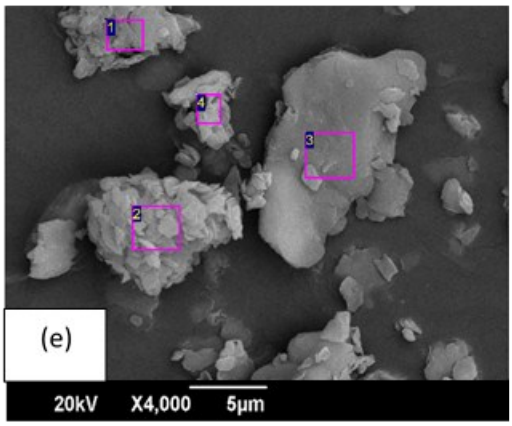
Figure S3 SEM micrographs (x2000) for: (a) r; (b) r-LD; (c) R; (d) R-LC; (e) R-BC; (f) RI and (g) RI-PC clay catalysts; purple-squares highlight surface zones where Fe concentration was determined by EDS for variance analysis.

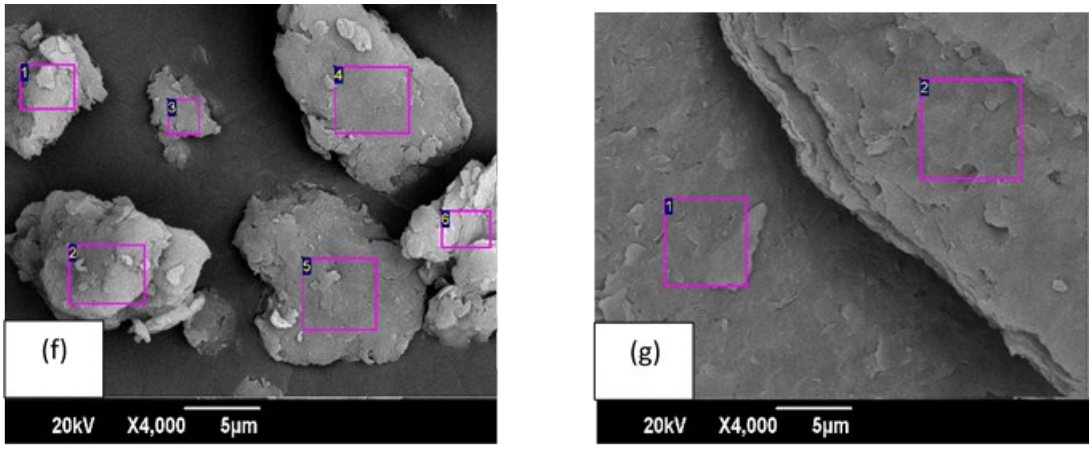

(a)
20kV X4,000 5μm


(b)
20kV X4,000 5μm


(c)
20kV X4,000 5μm


(d)
20kV X4,000 5μm


(e)
20kV X4,000 5μm



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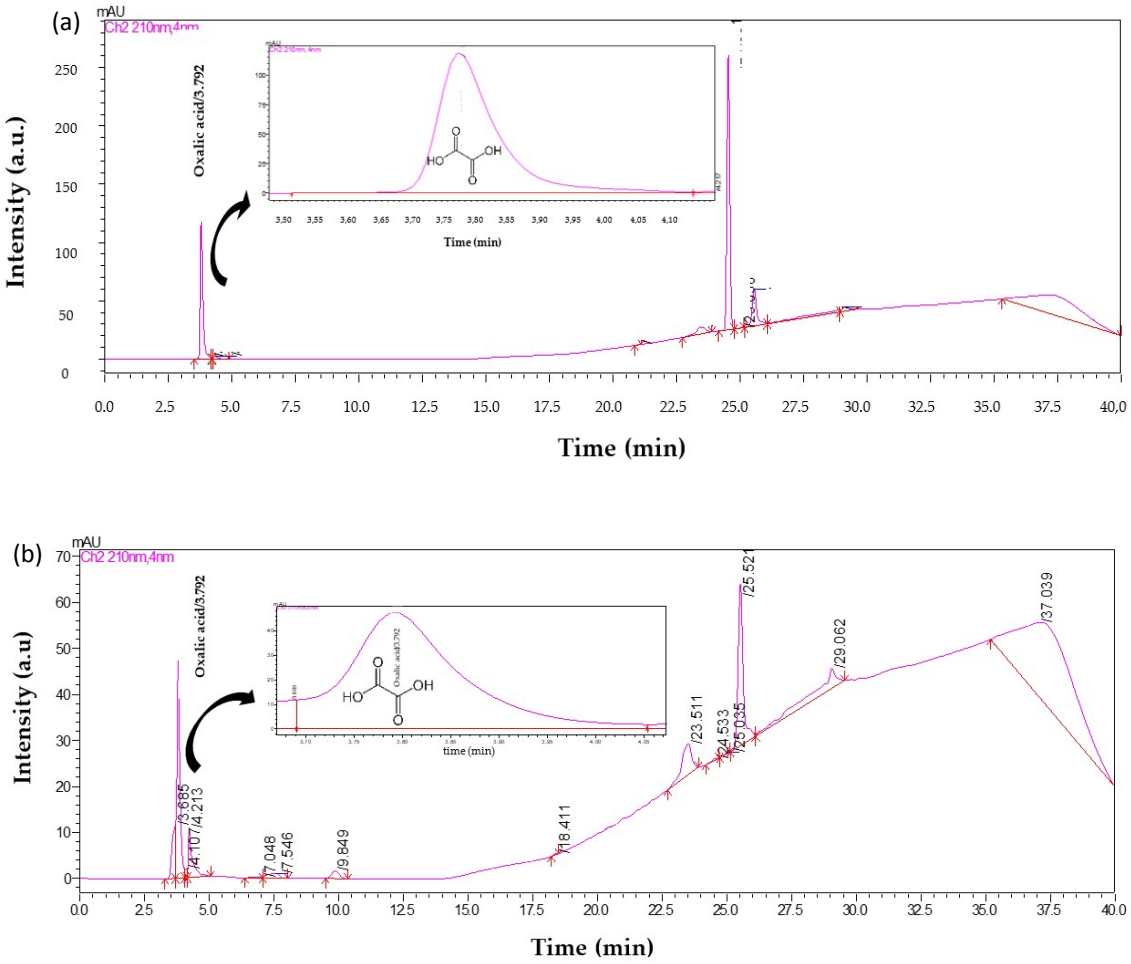

Figure S5 LC-DAD chromatograms of the (a) r-LD and (b) R-LC catalysts after 180 min of reaction (plus 30 min of pre-equilibrium period): comparison of diluted vs. concentrated precursors with oxalic acid detected.

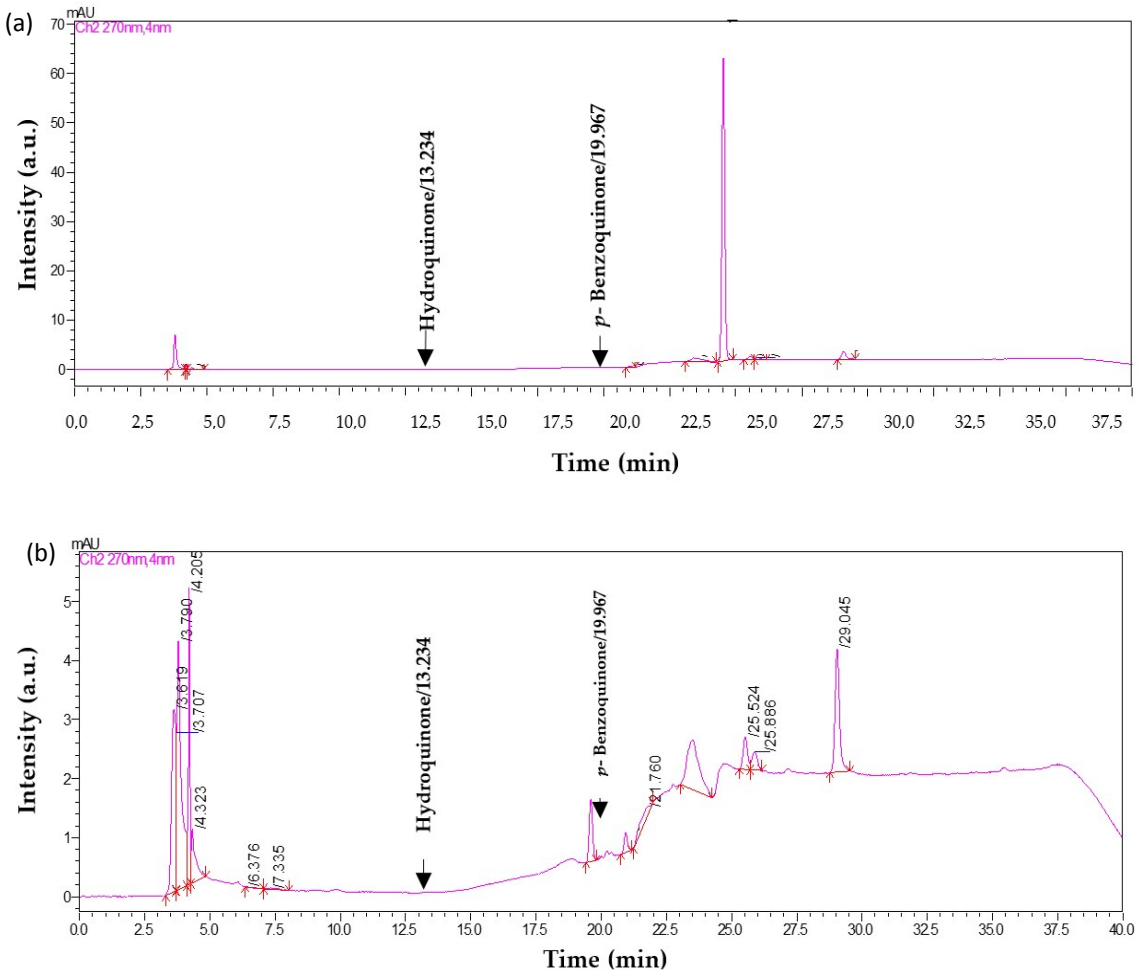

Figure S6 LC-DAD chromatograms of the (a) r-LD and (b) R-LC catalysts after 180 min of reaction (plus 30 min of pre-equilibrium period): comparison of diluted vs. concentrated precursors showing hydroquinone and *p*-benzoquinone expected retention times.

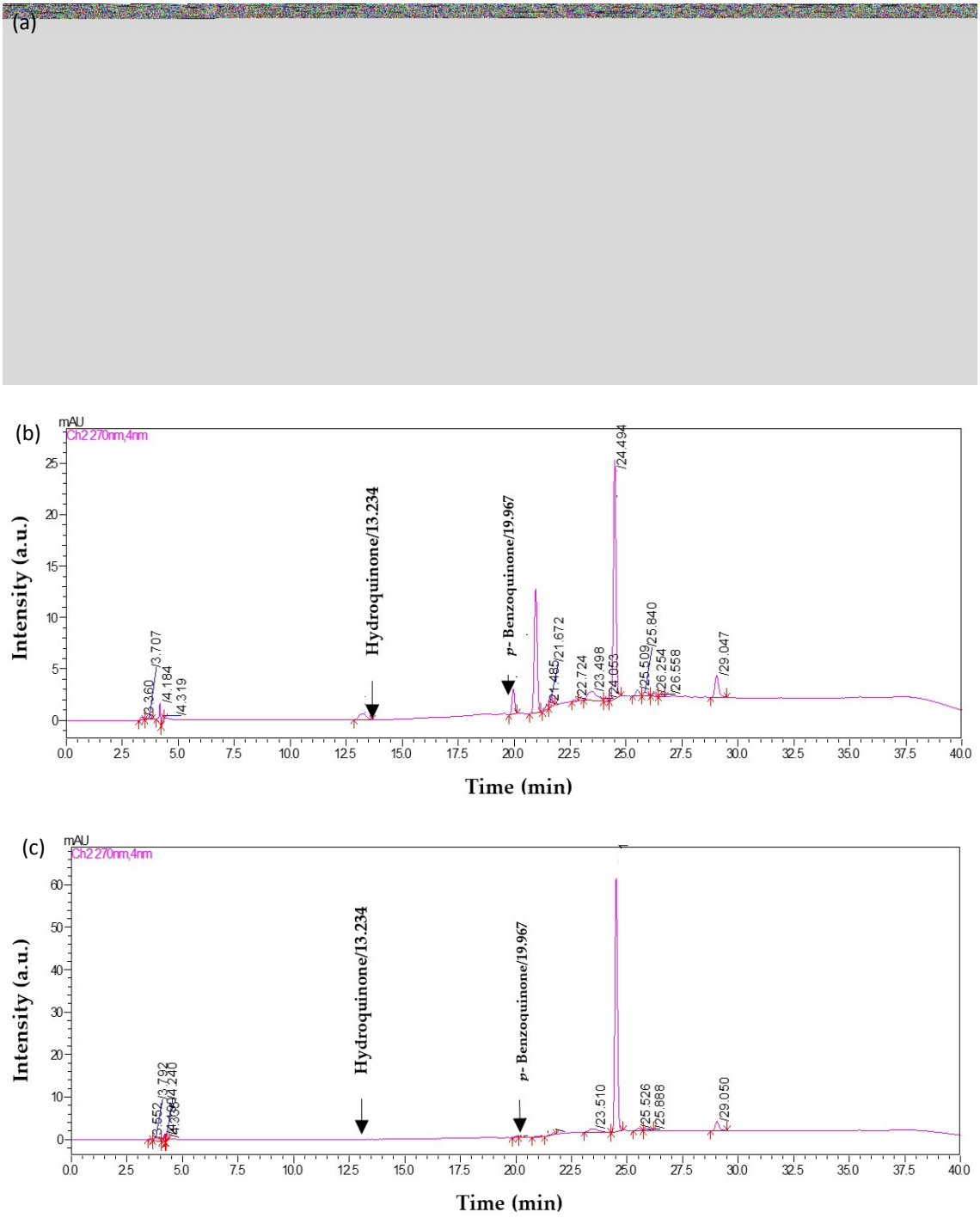

Figure S7 LC-DAD chromatograms of the (a) R-LC, (b) R-BC and (c) RI-PC catalysts after 180 min of reaction (plus 30 min of pre-equilibrium period): comparison lab- vs. bench- vs. pilot-scale preparations of the Al/Fe-PILC showing hydroquinone and *p*-benzoquinone expected retention times.