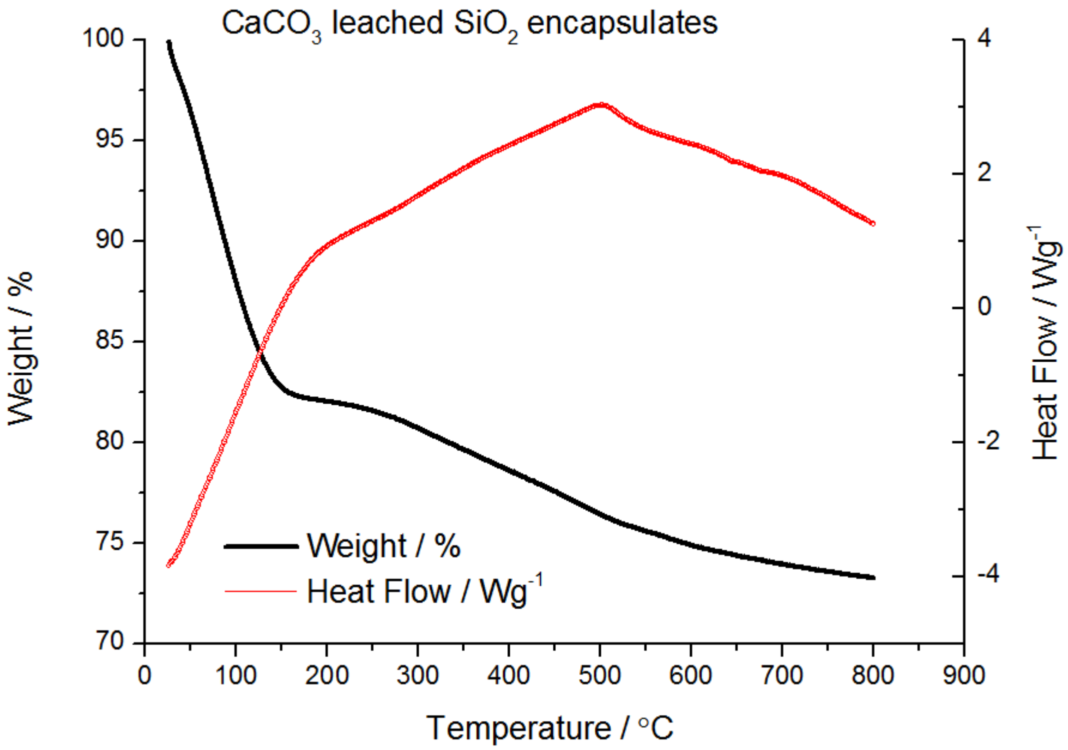
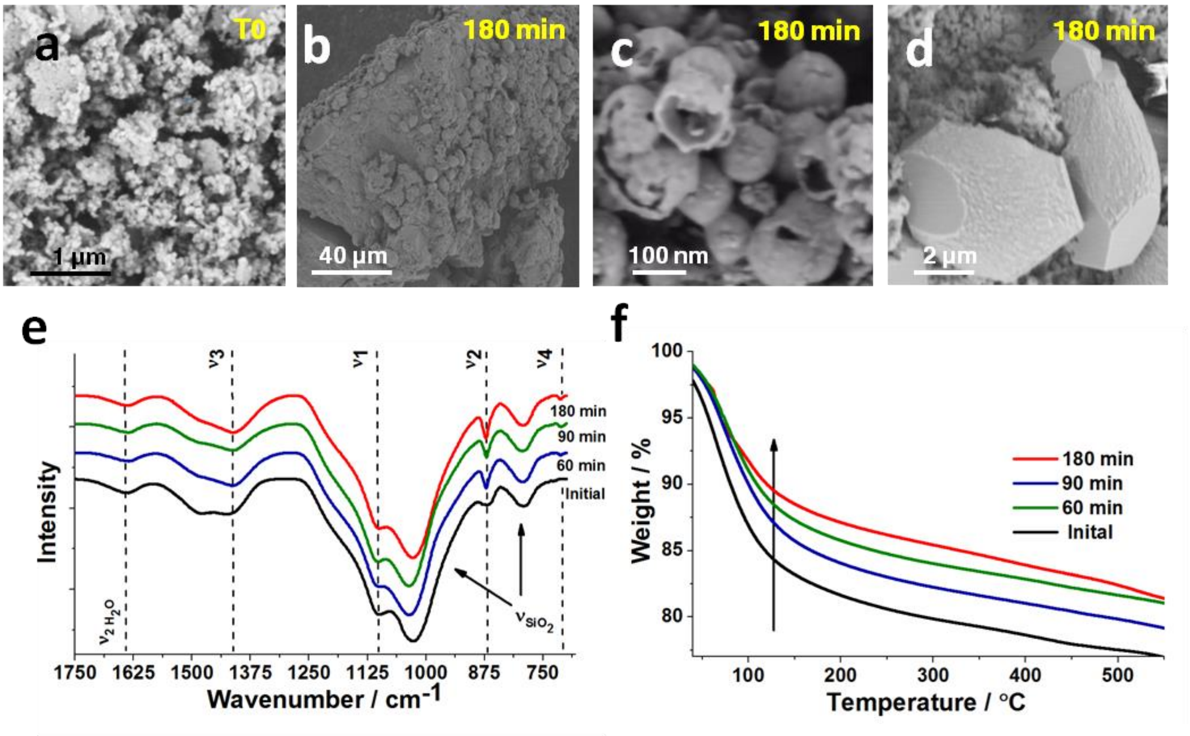
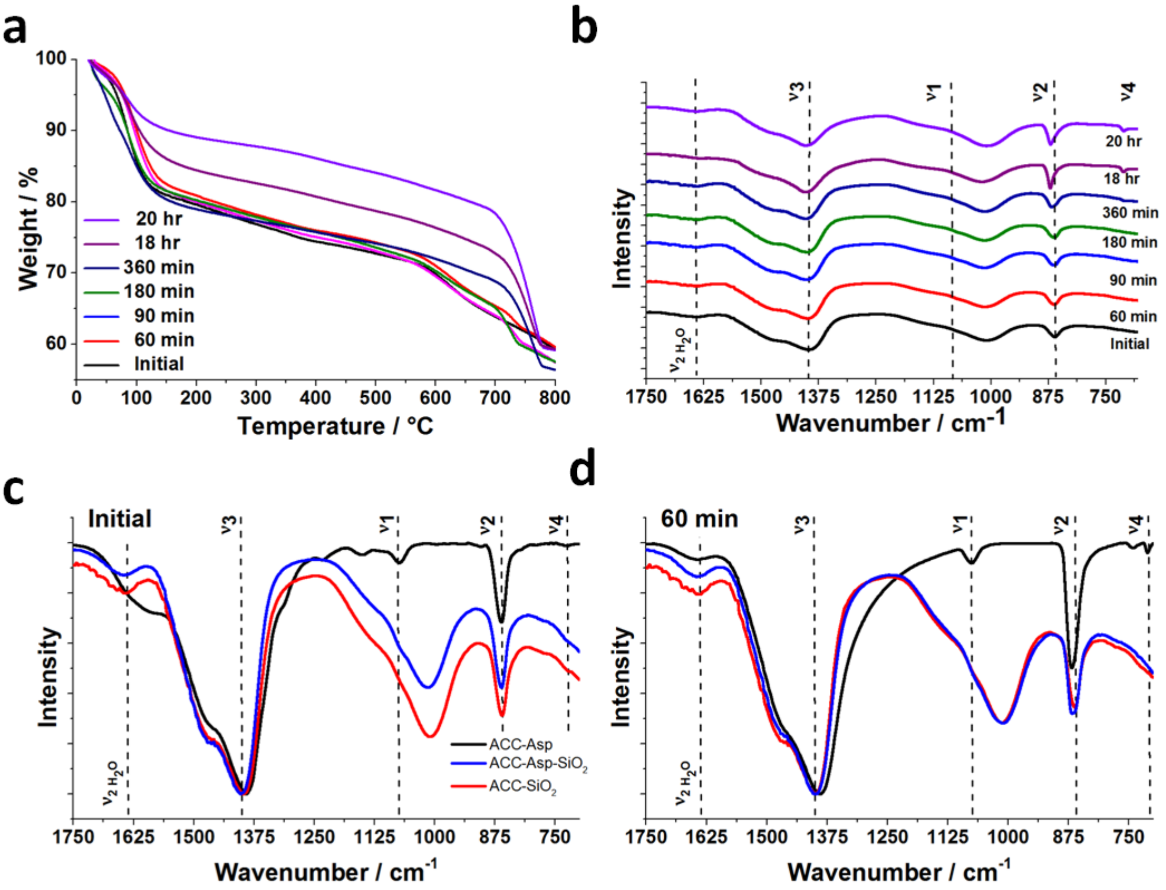


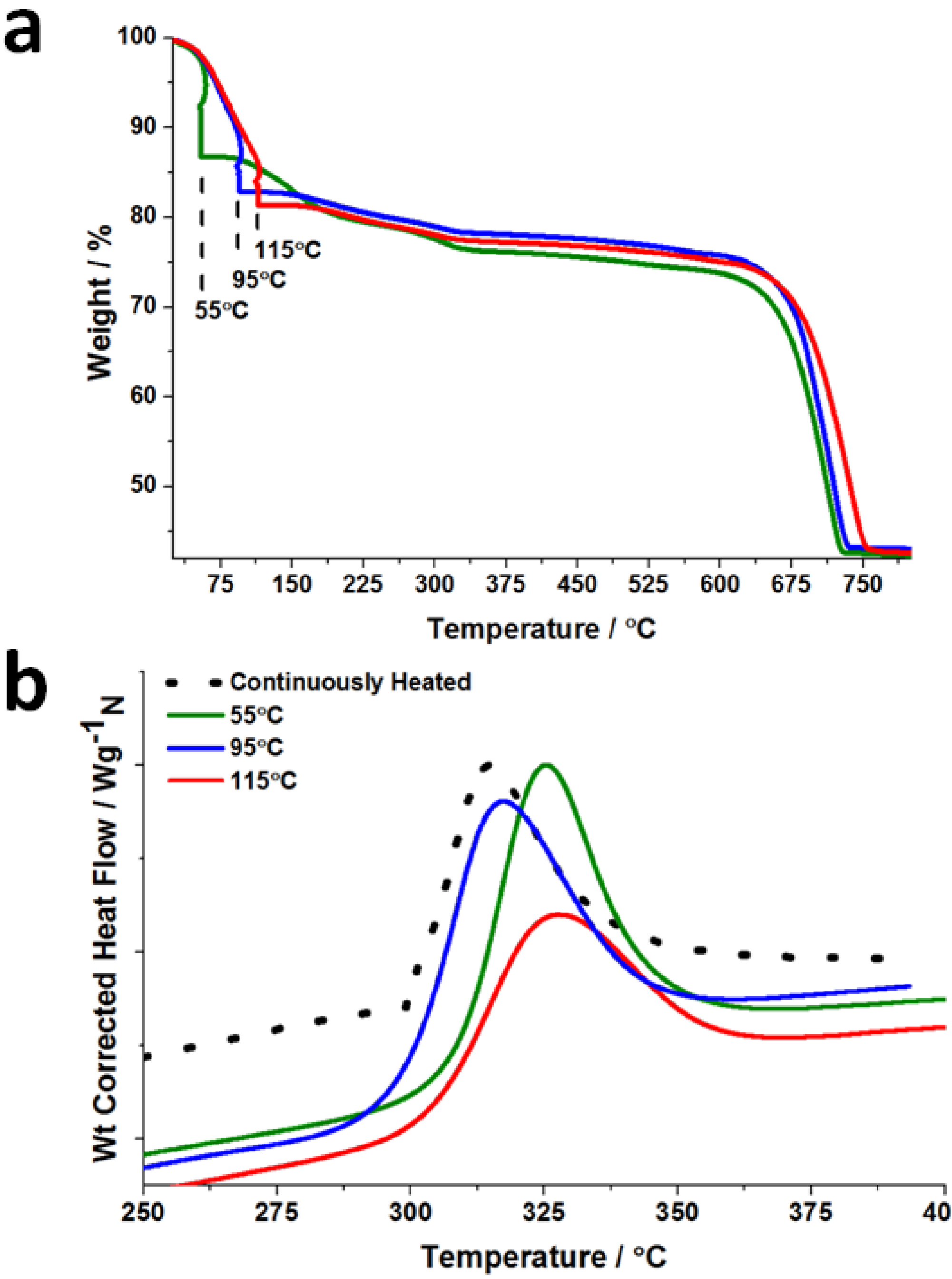
Supplementary Figure 1 Thermogravimetric analysis (TGA) of silica shells formed by leaching the $CaCO_3$ from ACC-SiO_2 particles by incubating the particles in HCl for 24 hours. TGA measurements were performed at a heating rate of 15°C /min under 100ml N_2/min.



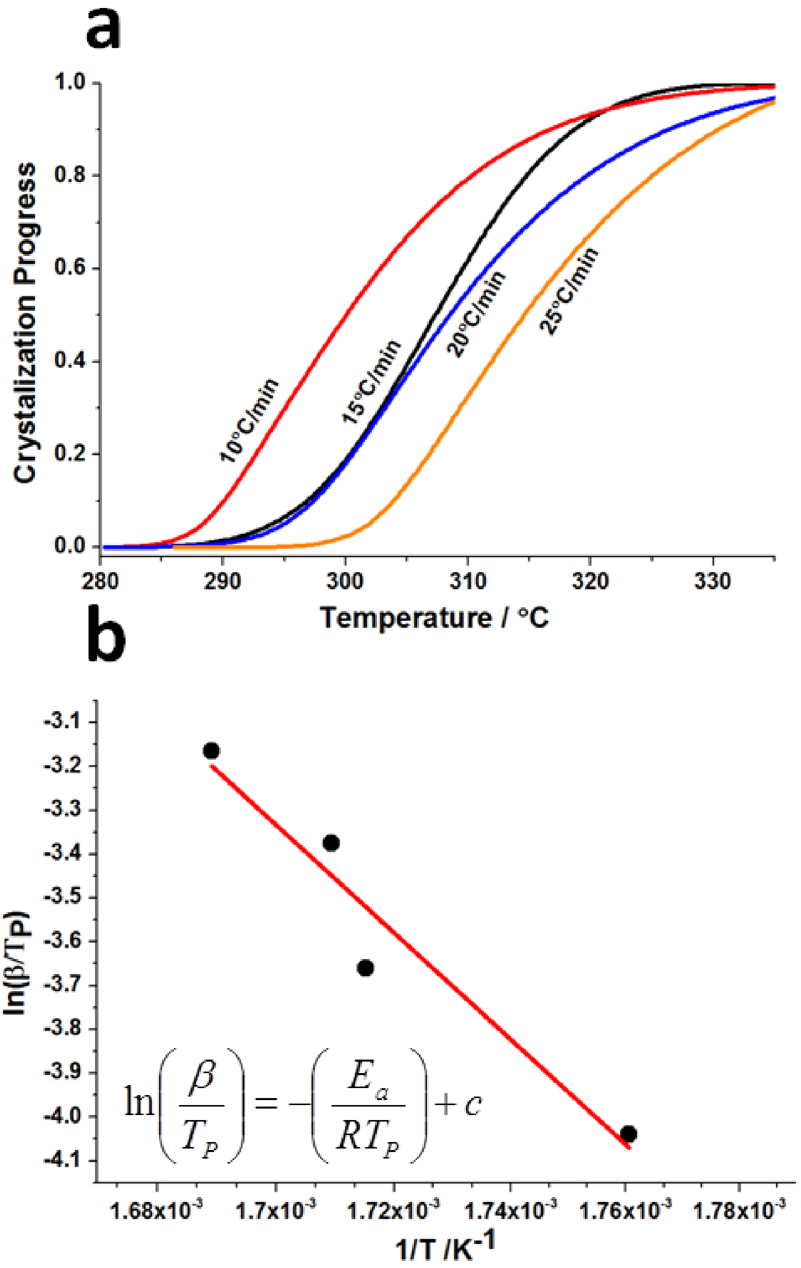
Supplementary Figure 2 Characterization of pure ACC particles coated with silica shells and their dehydration on incubation in solution with time. (a-d) SEM images show that the silica does not coat all ACC particles sufficiently to prevent rapid dissolution and transform to calcite. (a) Initially prepared particles are identical in appearance to ACC synthesised in the presence of NaSiO$_2$. (b) Silica-coated ACC samples after 180 minutes in solution, showing the presence of (c) empty silica shells and (d) calcite particles. These calcite particles give an apparent constant background in IR spectra. (e) IR spectra of coated ACC particles after different incubation times in solution, further indicate an increased SiO$_2$ presence in formed precipitate as compared with ACC coprecipitated with silica attributed to the higher concentration used.[36] (f) TGA also demonstrates similar dehydration behaviour of the silica-coated pure ACC particles as ACC particles co-precipitated with silica.



Supplementary Figure 3 Transformation of ACC-Asp-SiO_2 particles prepared by combining equal volumes of 10 mM Na_2CO_3/ 6 mM Na_2SiO_3 and 10 mM $CaCl_2$ $2H_2O$/ 5 mM aspartic acid. (a) Thermogravimetric analyses and (b) IR spectra of ACC-Asp-SiO_2 after different incubation times in solution. The weight loss during heating decreases with time spent in solution because progressively more dehydration has already occurred in solution. The small band at ≈ 714 cm^{-1} corresponding to crystalline $CaCO_3$ was observed in IR spectra after 18 hours, with the first hint of crystallinity appearing after ≈ 5 hours. (c and d) Comparison of IR spectra of ACC-SiO_2 (—), ACC-Asp (—) and ACC-Asp-SiO_2 (—) particles after incubation in solution for (c) 0 mins and (d) 60 mins. The spectra show the development of bands for vaterite and calcite at ≈ 714 cm^{-1} and 747 cm^{-1} for the ACC-Asp particles after 60 mins, while these are not observed for the coated ACC particles.



Supplementary Figure 4 Full temperature range TGA (a) and limited DSC (b) of uncoated ACC particles. ACC particles were produced by combining equal volumes (0.5-1.5 ml) of 1 M $(NH_4)_2CO_3$ (pH 9.15) with 1 M $CaCl_2$ (pH ~6.8) at 4°C. A heating rate of 15°C/min with a single isothermal annealing period (for 100min at 55, 95 or 115°C) under a constant gas flow rate of 100ml N_2/min was used. Note from (b) that no consistent variation in the position of the crystallization peak was found.



Supplementary Figure 5 Evaluation of the Progress of ACC Crystallization. (a) Curves showing the crystallisation progress of uncoated ACC samples under heating with an intermediate annealing period (135°C for 100min), as derived from DSC scans performed with applied heating rates of 10, 15, 20 and 25^{o}C/min, under 100ml N_2/min. The progress of crystallisation, 0-1, was obtained by integration of the crystallisation associated exothermic peak present in obtained DSC scan. (b) Corresponding Boswell plot[44] required for the calculation of activation energies associated with crystallisation, E_a. β = heating rate, T_P = DSC central peak temperature.

Supplementary Table 1 Summary of the intensities of FTIR peaks of uncoated ACC and ACC-SiO_2 particles at different stages of dehydration with respect to temperature or time. α represents the degree of dehydration from 0-1, $CaCO_3$: x H_2O the assumed molecular composition and Wt% H_2O the total sample, water weight percentage. Spectra were normalized with respect to the right hand side of the v_3 peak at 1402 cm^{-1} (v_{3R}).

ACC after isothermal annealing

α	$CaCO_3$: x H_2O	Wt% H_2O	Temperature [°C]	v_3/v_{2H2O}	v_3/v_{3R}	v_3/v_1	v_3/v_2	v_3/v_4	v_3/v_{SiO2}
0.00	1.40	20.00	25	6.67	1.22	1.69	2.78	46.54	-
0.49	0.71	10.20	50	9.09	1.20	1.82	2.50	50.76	-
0.78	0.31	4.40	100	10.00	1.18	2.50	2.33	47.65	-
0.98	0.03	0.40	180	11.11	1.32	3.75	2.08	49.87	-
0.99	0.01	0.20	200	14.29	1.20	4.52	2.00	37.50	-
1.00	0.00	0.00	250	16.67	1.23	4.52	2.00	11.11	-

ACC-SiO_2 in solution

α	$CaCO_3$: x H_2O	Wt% H_2O	Time [min]	v_3/v_{2H2O}	v_3/ v_{3R}	v_3/v_1	v_3/v_2	v_3/v_4	v_3/v_{SiO2}
0.00	1.40	20.00	0	11.29	1.30	2.50	2.94	45.33	1.53
0.21	1.11	15.80	30	11.11	1.33	2.63	2.70	50.00	1.59
0.39	0.85	12.20	60	11.11	1.28	2.78	2.38	43.47	1.64
0.54	0.64	9.20	90	12.50	1.37	2.78	2.04	51.37	1.75
0.67	0.46	6.60	120	12.50	1.41	2.94	1.82	50.98	1.86
0.76	0.34	4.80	150	16.67	1.37	5.00	1.85	36.33	2.28
0.98	0.03	0.40	500	25.00	1.30	5.26	1.85	12.50	2.38