Supplementary Material:

Experimental determination of impure CO_2 alteration of calcite cemented cap-rock, and long term predictions of cap-rock reactivity

J.K. Pearce and G.K.W. Dawson

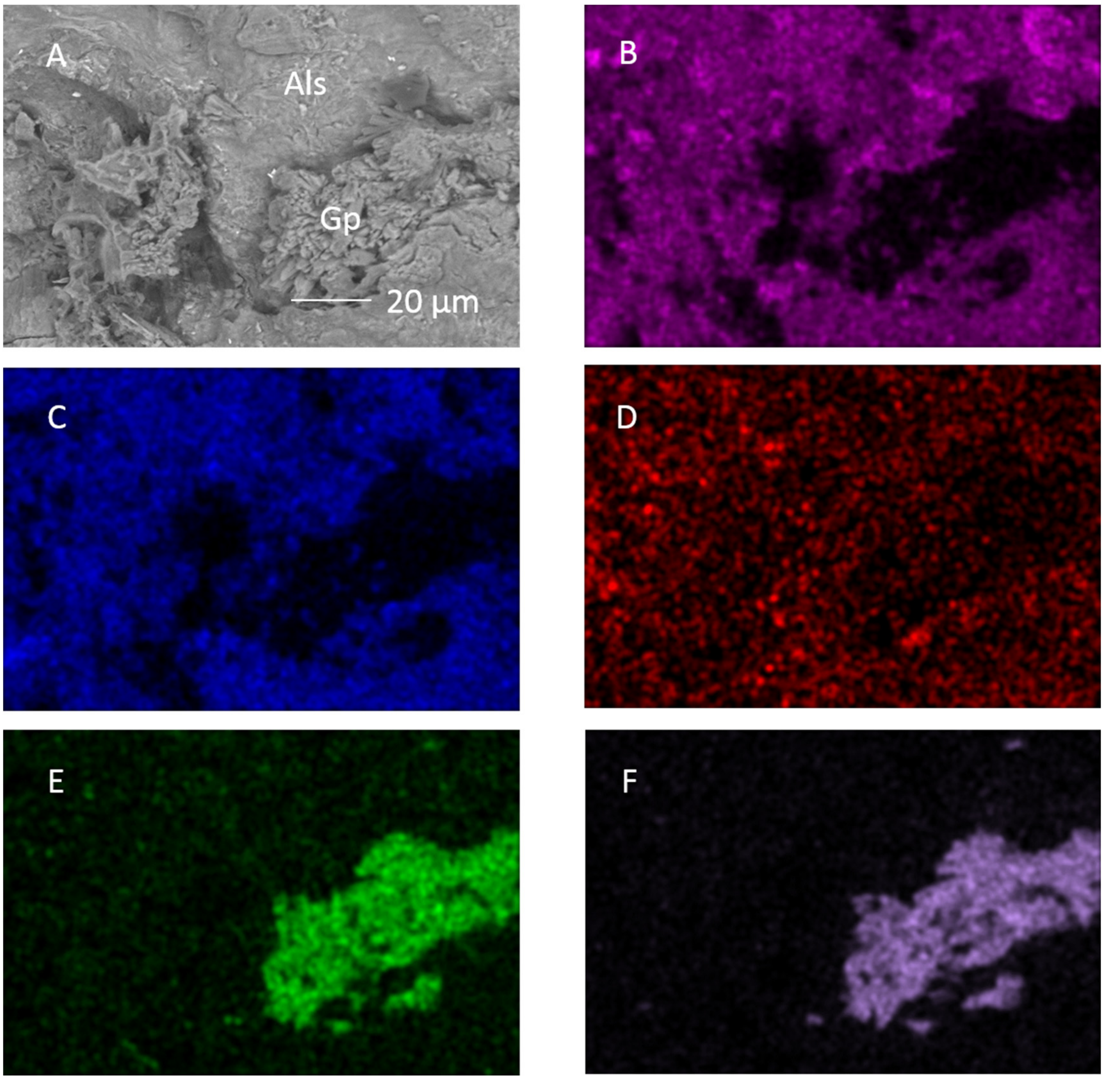

Figure S1. Scanning Electron microscope (SEM) image and EDS element maps after reaction showing precipitated gypsum (Gp), Aluminosilicate layer (Als) and skeletal clay (indicated with an arrow). Colours in B - F indicate a relatively stronger element concentration. A) SEM image, B) Pink = Si, C) Blue = Al, D) Red = Fe, E) Green = Ca, F) Purple = S.

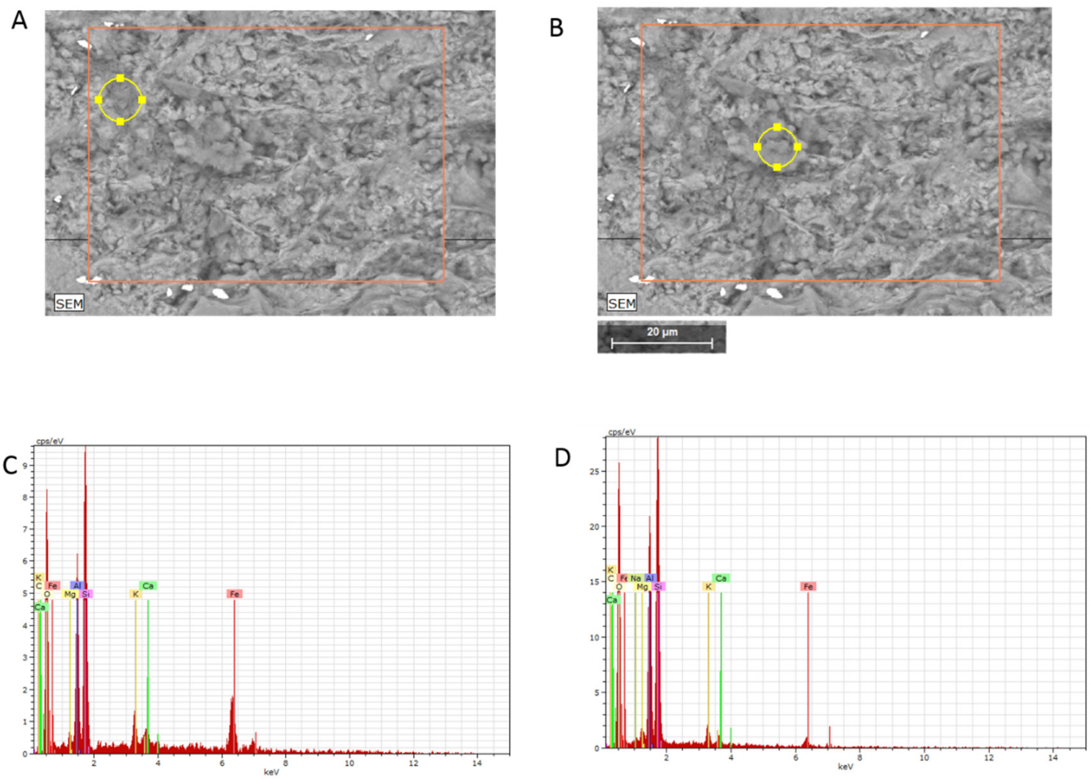

Figure S2. SEM and EDS spectra of corroded chlorite surface after reaction. A) SEM image of corroded chlorite surface, B) Circle marks a precipitated amorphous material. C) EDS spectrum of corroded chlorite surface marked in A, D) EDS spectrum of precipitated amorphous material marked in B.

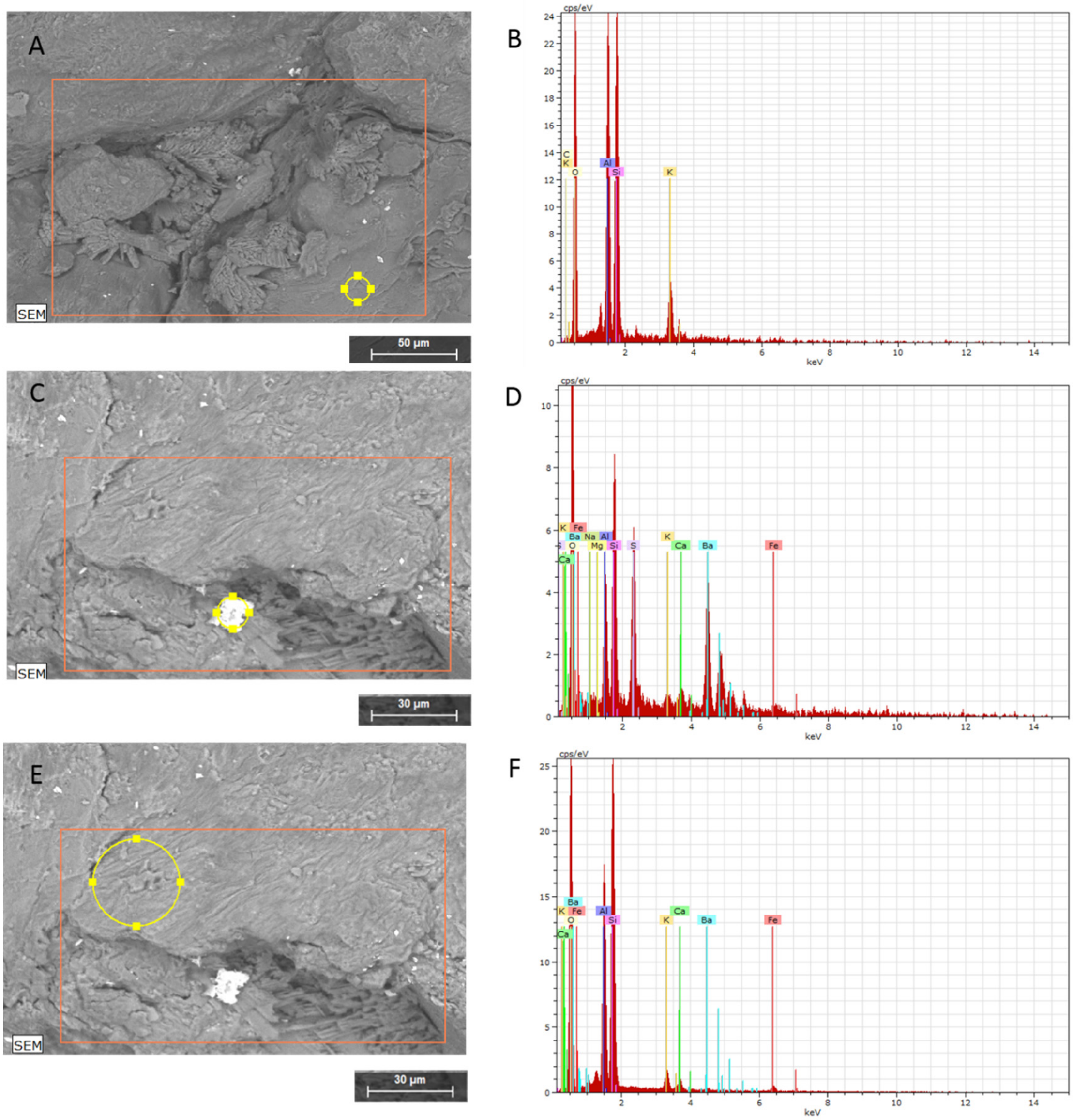

Figure S3. SEM images and EDS spectra of the inside surface of the block after reaction. A) Gypsum growing in created pore, B) EDS spectrum of marked K-feldspar grain in A, C) Precipitated barite, D) EDS spectrum of barite in C, E) precipitated aluminosilicate material, F) EDS spectrum of precipitate marked in E with a Si:Al ratio similar to kaolinite.

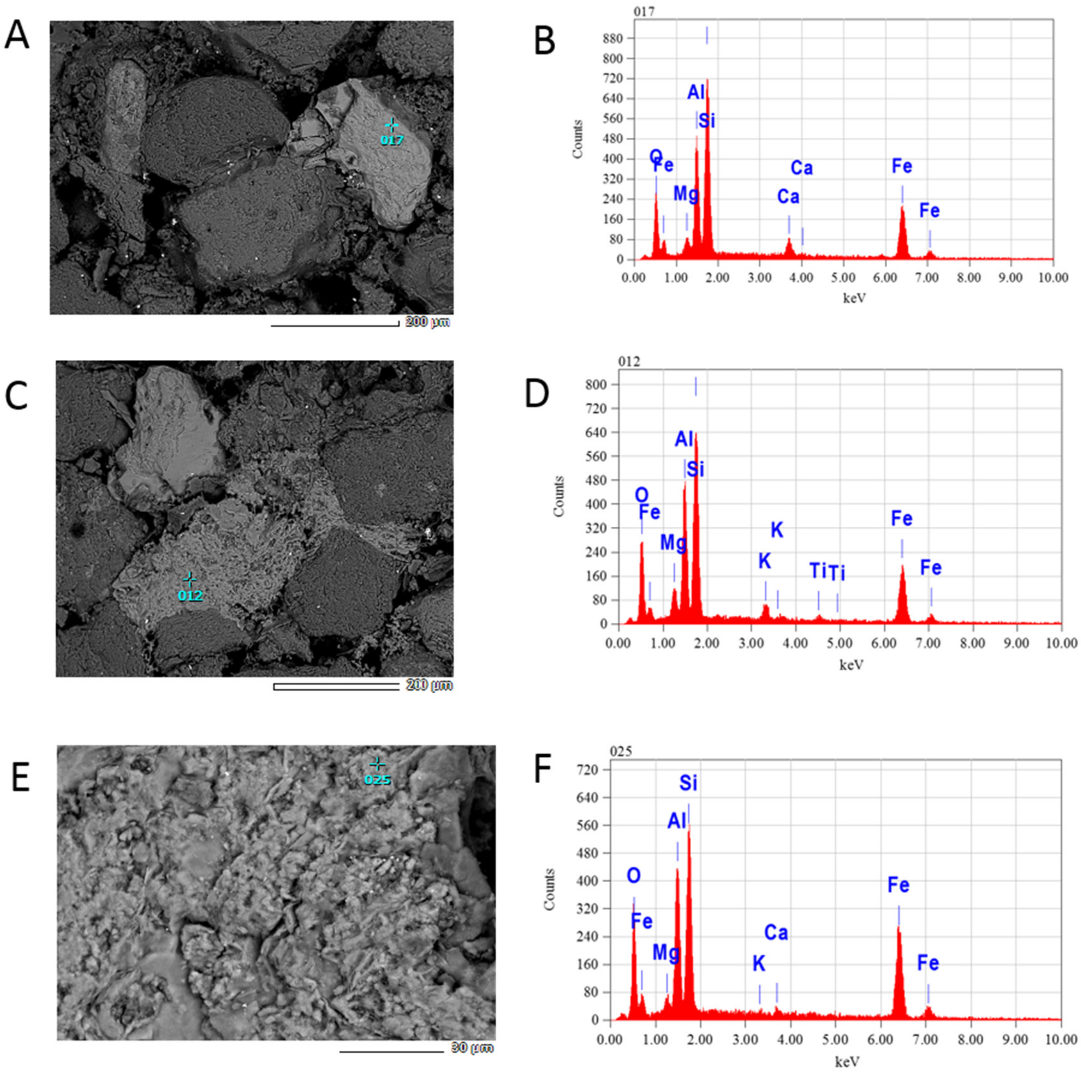

Figure S4. SEM images and EDS spectra of the exposed block surface after reaction. A) and B garnet grain appears unaltered, C and D) mica appears unaltered, E and F) chlorite.

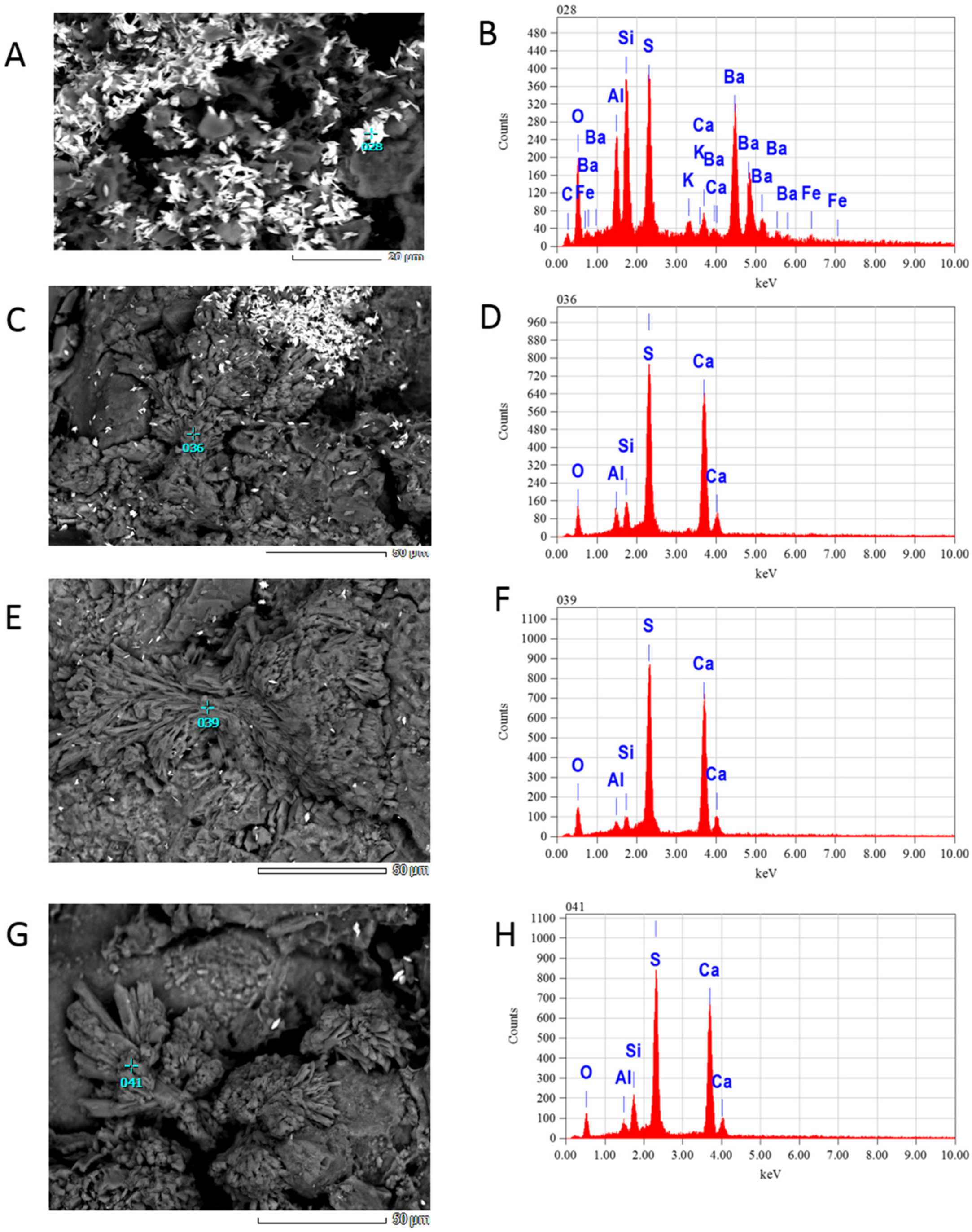

Figure S5. SEM images and EDS spectra of precipitated material after reaction: A) and B) Bright barite crystals precipitated on gypsum, C) to H) Gypsum growing from the pore space created by calcite dissolution.

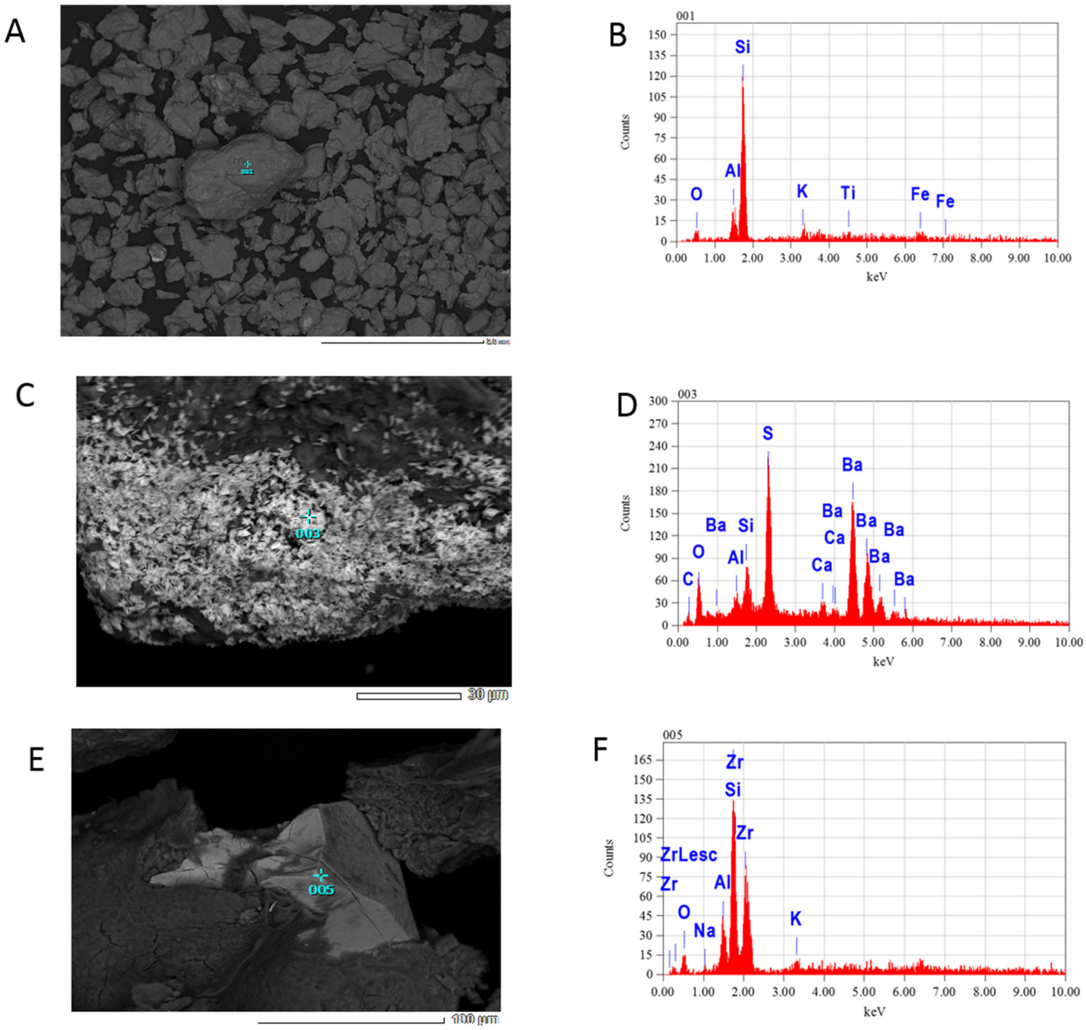

Figure S6. SEM images and EDS spectra of disaggregated grains after reaction recovered from the reactor: A) and B) Framework grains covered in Fe-oxide or aluminosilicate coatings, C) and D) Precipitated bright barite crystals, E) and F) zircon with amorphous precipitated material coating surrounding grains.

Table S1. Geochemical modelling parameters used for all models, where K is a rate constant, Ea and activation energy, n is a power term, the pre-exponential nucleation factor mod indicates a modified parameter used for modelling the experimental data. Refer to the main manuscript text for source references.

Mineral	$K25_{(acid)}$ $mol/cm^2/s$	$Ea_{(acid)}$ kJ/mol	n	$K25_{(neut)}$ $mol/cm^2/s$	$Ea_{(neut)}$ kJ/mol	$K_{(precip)}$ $mol/cm^2/s$		mod
Quartz				1.70E-17	68.7	K(diss)	2.00E+10	
K-feldspar	8.71E-15	51.7	0.5	3.89E-17	38	K(diss)	2.00E+10	
Albite	6.92E-15	65	0.457	2.75E-17	69.8	K(diss)	2.00E+10	
Andesine	1.32E-13	53.5	0.54	3.39E-16	57.4	K(diss)	2.00E+10	
Kaolinite	4.90E-16	65.9	0.777	6.61E-18	22.2	K(diss)/10	2.00E+10	2.00E+11
Illite/muscovite	1.91E-16	46	0.6	8.91E-20	14	K(diss)	2.00E+10	
Smectite	1.05E-15	23.6	0.34	1.66E-17	35	K(diss)	2.00E+10	
Biotite	1.45E-14	22	0.525	2.82E-17	22	K(diss)	2.00E+10	
Fe-Mg-Chlorite	1.62E-14	25.1	0.49	1.00E-17	94.3	K(diss)	2.00E+10	
Calcite	5.01E-05	14.4	1	1.55E-10	23.5	K(diss)	1.00E+10	
Ankerite	1.59E-08	45	0.9	1.26E-13	62.76	K(diss)/1e5	3.00E+10	
Siderite	1.59E-08	45	0.9	1.26E-13	62.76	K(diss)	2.00E+10	
Fe-oxide	4.07E-14	66.2	1	2.51E-19	66.2	K(diss)	1.00E+10	9.00E+10

Table S2. Porosities (%) used for cap-rock models [24]. WW1 = West Wandoan 1 well, Cab1 = Cabawin 1 well. CalCem = Calcite cemented, MudS = mudstone, SidCem = siderite cemented.

WW1 CalCem	WW1 MudS	Cab1 SidCem	Cab1 Shale
8	9	10	10

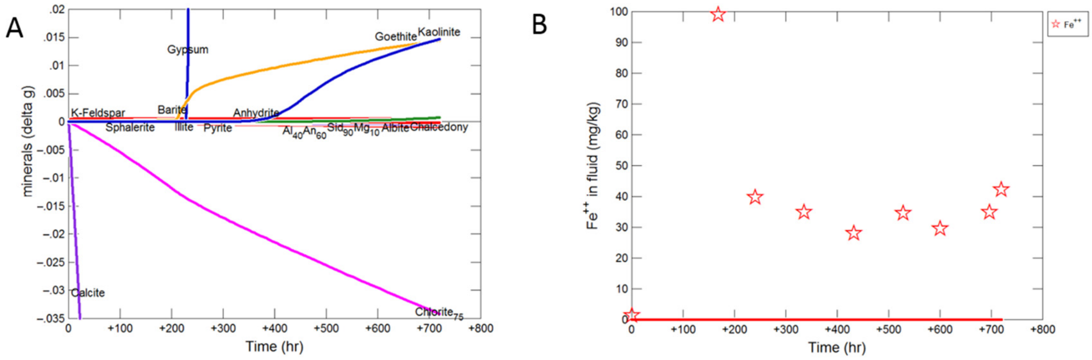

Figure S7. Experimental models (lines) and experimental data (symbols) for calcite cemented cap-rock reaction: A) Magnified view of minor mineral mass (grams) dissolving and precipitating in the final experimental model. B) Dissolved Fe when hematite precipitation was unsuppressed.

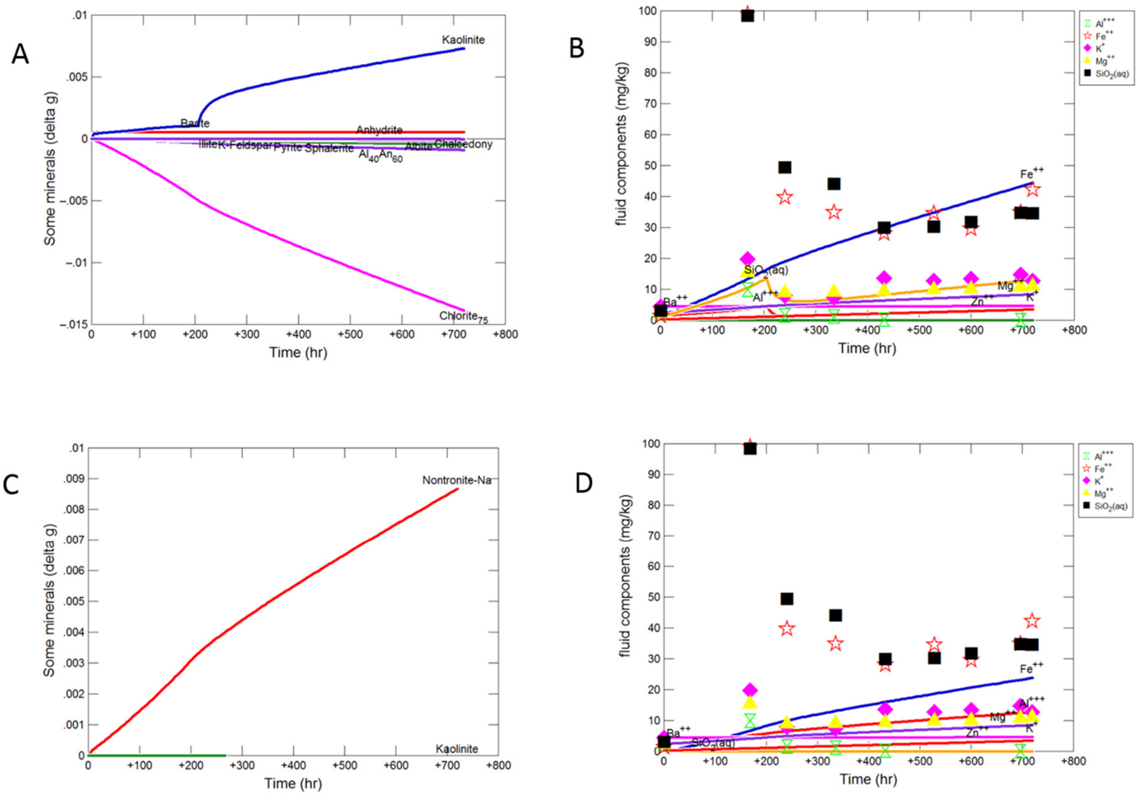

Figure S8. Additional experimental models (lines) and experimental data (symbols) for calcite cemented cap-rock reaction: A) and B) change in minor minerals mass (grams) and water chemistry without kaolinite and goethite script file modification, C) and D) smectite and kaolinite mineral precipitation mass (grams), and water chemistry when smectite precipitation was unsuppressed.