Electronic Supplementary information for:

Addressing the Stiffness-Toughness Conflict in Hybrid Double Network Hydrogels through a Design of Experiments Approach

Vinay Kopnar [a], Laurie Carlyle [b], Emerald Liu [b], Suchet Khaenyook [c], Adam O'Connell [d], Natasha Shirshova [b] and Anders Aufderhorst-Roberts [a,e]

[a] Department of Physics, Durham University, Lower Mountjoy, South Rd, Durham DH1 3LE, UK

[b] Department of Engineering, Durham University, Lower Mountjoy, South Rd, Durham DH1 3LE, UK

[c] Reckitt (SSL Manufacturing (Thailand) Ltd.), 100 Moo 5, Bang Na Trad Road, KM36, Bangsamak, Bangpakong, Chachoengsao, 24180 Thailand

[d] Polymer Science Platform, Reckitt Benckiser Health Care UK Ltd, Dansom Lane S, Hull HU8 7DS, UK

[e] School of Engineering, University of Liverpool, Liverpool L69 3GH, UK.; E-mail: anders.aufderhorst-roberts@liverpool.ac.uk

S1: A pair of datasets do exist where only CMBA is increased from 0.25mM (L in Table 1 from the manuscript) to 0.5mM (J' in Table 3 from the manuscript) while the rest of the factors are kept constant. For 0.25mM, the measured values of Young's modulus (14.2 kPa and 16.8 kPa) falls close to the lower limit of the confidence interval of prediction (16.1 kPa). Meanwhile at CMBA = 0.5mM, the measured value (32.7 kPa) corresponds closely to the upper limit of the corresponding confidence interval (33.5 kPa). This would indicate that increasing the limit on CMBA has very little effect on the other factors.

5-parameter FE model performance summary

R^2	R^2_{adj}	R^2_{pred}	5-fold R^2
90.98%	88.72%	83.19%	84.45%

S2: Performance of model describing FE. 5-fold cross-validation was carried out to test the effect of overfitting. To carry this out, the data was split into 5 equal folds and the model is trained and tested 5 times, each time using a different fold as the test set and the remaining four folds as the training set. 5-fold R^2 presents the average of the different R^2 from the 5 iterations. 5-fold R^2 is closer R^2 highlighting the effect of overfitting is minor.
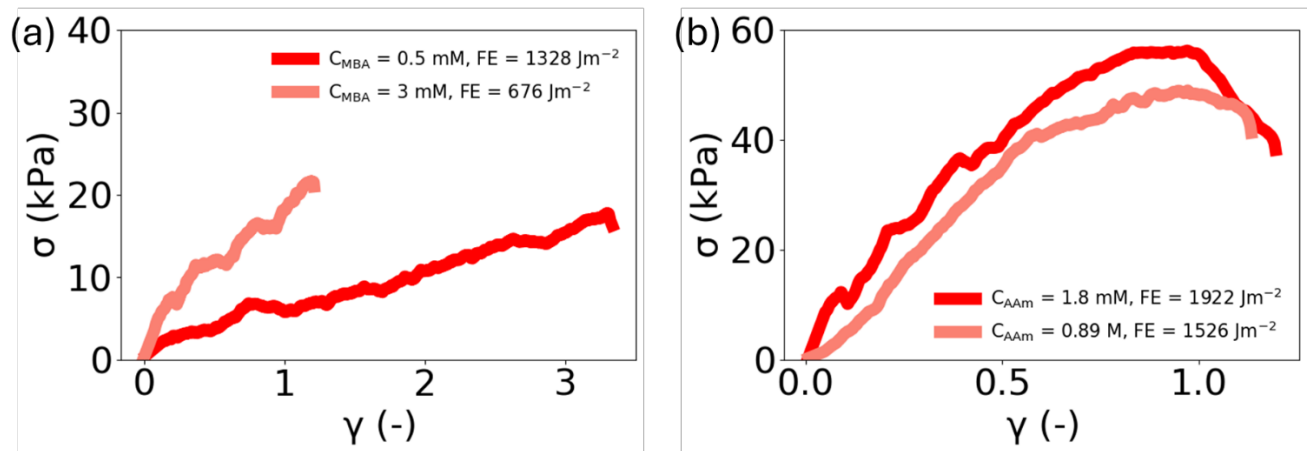
S3: Stress-strain curves to demonstrate the effect of increasing MBA and AAm concentration on FE. (a) shows the effect of C_{MBA} on FE as C_{MBA} is increased from 0.5 mM to 3 mM. Concentration of all other parameters were kept constant (C_{Alg} = 1.6 %w/v, C_{CaCO_3} = 0.011 M, C_{APS} = 1.1 mM, C_{AAm} = 1.8

M); (b) shows the effect of C_{AAm} on FE as C_{AAm} is increased from 0.89 mM to 1.8 mM. Concentration of all other parameters were kept constant (C_{Alg} = 1.6 %w/v, C_{CaCO_3} = 0.045 M, C_{APS} = 0.62 mM, C_{MBA} = 3 mM)
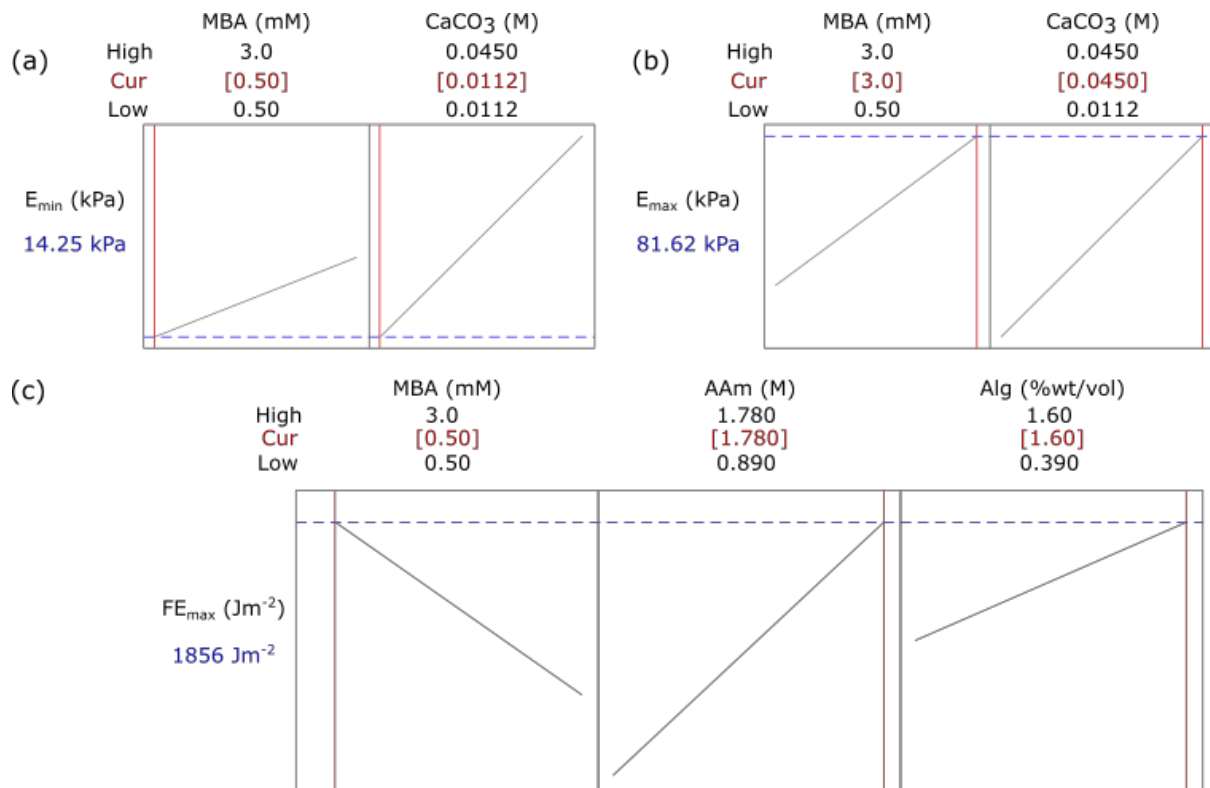
S4: Optimization Plots for finding (a) minimum E (E_{min}), (b) maximum E (E_{max}), and (c) maximum FE (FE_{max}). They demonstrate the effect of each factor (column) on the responses. The vertical red lines on the graph depict the current factor settings. The numbers at the top of each column represent the current factor level settings (in red). The horizontal blue lines and numbers represent the responses at the current factor level.

Serial number	MBA (mM)	CaCO3 (M)	AAm (M)	Alg (%w/v)	APS (mM)
1	2.72	0.018	1.09	1.4	0.28
2	0.25	0.011	0.89	1.6	1.1
3	3	0.011	1.7	0.4	0.2
4	3	0.016	0.96	1.58	1.1
5	3	0.035	0.89	1.12	1.1
6	2.97	0.04	0.9	1.57	1.05
7	0.52	0.019	1.76	0.39	1.07
8	3	0.018	1.69	0.8	0.28
9	0.36	0.045	1.76	0.39	1.1

S5: 9 formulations explored for validation of the models