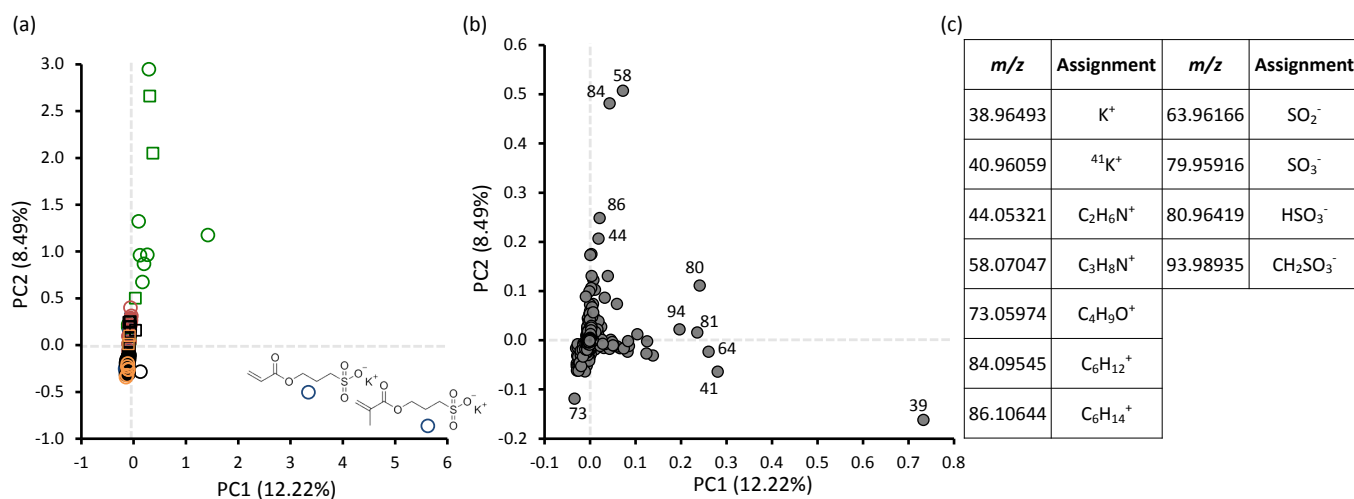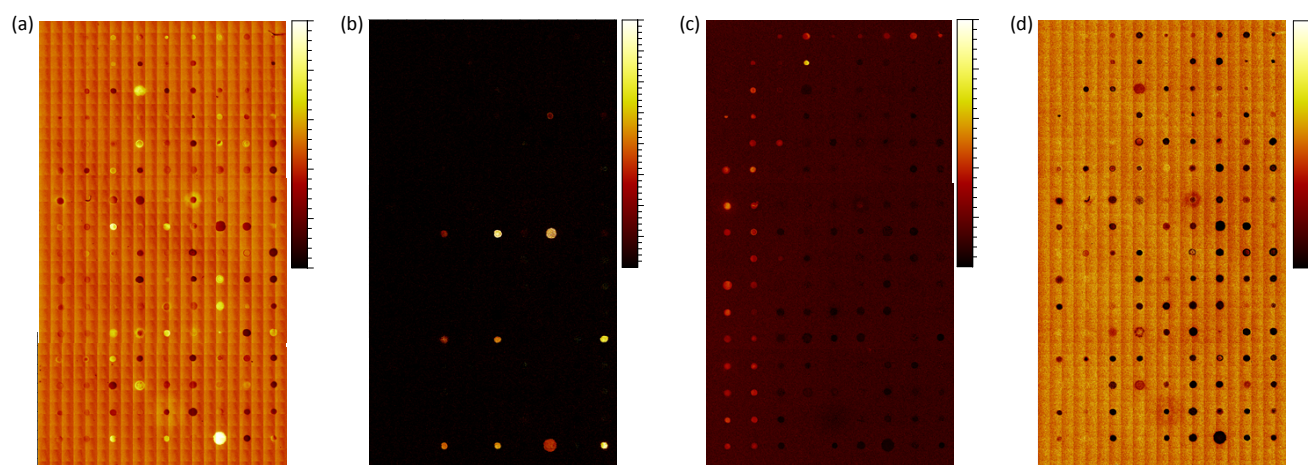**Supporting Figure S1.** a) PLS WCA model consisting of a training set of 103 samples (●) and a test set of the remaining 34 samples (○), root-mean-square error of cross validation (RMSECV) curve used to determine the number of latent variables for PLS regression analysis of ToF-SIMS and WCA datasets (inset). b) PLS cell adhesion model consisting of a training set of 107 samples (○) and a test set of the remaining 34 samples (●), root-mean-square error of cross validation (RMSECV) curve used to determine the number of latent variables for PLS regression analysis of ToF-SIMS and cell adhesion datasets (inset).


m/z	Assignment	m/z	Assignment
38.96493	K^+	63.96166	SO_2^-
40.96059	$^{41}K^+$	79.95916	SO_3^-
44.05321	$C_2H_6N^+$	80.96419	HSO_3^-
58.07047	$C_3H_8N^+$	93.98935	$CH_2SO_3^-$
73.05974	$C_4H_9O^+$		
84.09545	$C_6H_{12}^+$		
86.10644	$C_6H_{14}^+$		

Supporting Figure S2. PCA analysis of the mean-centered and square root mean scaled ToF-SIMS dataset a) Scores plot of PC1 versus PC 2. Chemical variance of two monomers within the 141 member library was identified. Sample groupings follow the same legend as in Figure 2. (b) Loadings plot of PC1 versus PC2. c) Ion structural assignments for loadings of largest variance for PC1 and PC2.
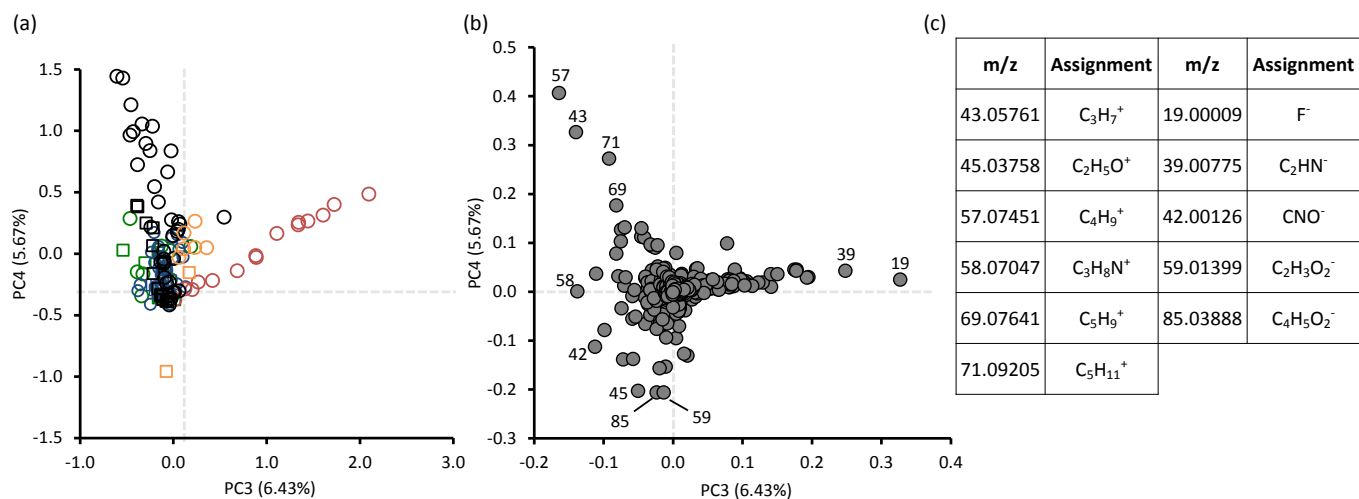
Supporting Figure S3. a) Scores plot of PC3 versus PC 4. Sample groupings follow the same legend as in Figure 2. (b) Loadings plot of PC3 versus PC4. c) Ion structural assignments for loadings of largest variance for PC3 and PC4.

The table in panel (c):

m/z	Assignment	m/z	Assignment
43.05761	$C_3H_7^+$	19.00009	F^-
45.03758	$C_2H_5O^+$	39.00775	C_2HN^-
57.07451	$C_4H_9^+$	42.00126	CNO^-
58.07047	$C_3H_8N^+$	59.01399	$C_2H_3O_2^-$
69.07641	$C_5H_9^+$	85.03888	$C_4H_5O_2^-$
71.09205	$C_5H_{11}^+$		

Supporting Figure S4. ToF-SIMS imaging of (a) total ion (negative), (b) ^+CF, (c) ^-CN and (d) $^-C_2H_5O_2$ (pHEMA ion) revealed no chemical spreading had occurred prior to the UV *in situ* polymerisation.
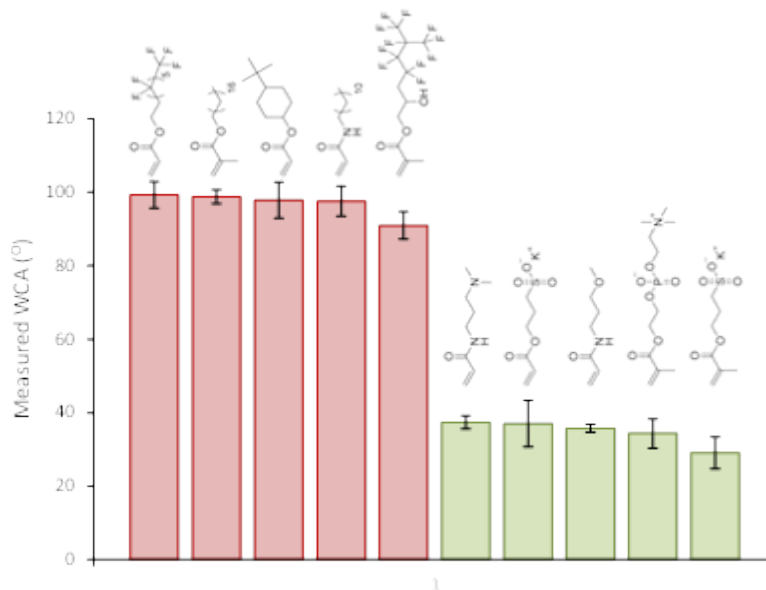
Supporting Figure S5. Five most hydrophobic (red) and hydrophilic (green) materials within the polymer microarray as measured by the WCA, standard deviation errors bars are determined from three replicates.
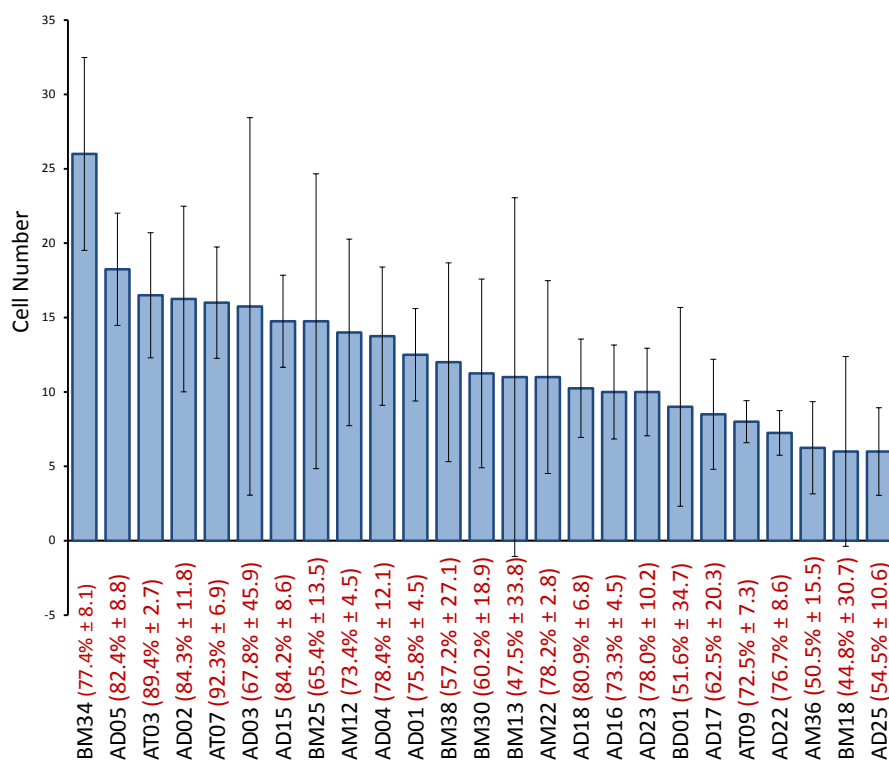

Supporting Figure S6. Top 25 materials ranked by Oct4 positive cell number, standard deviation error bars determined from four replicates. Percentage of Oct4 positive cells adhered to each material is displayed in brackets.
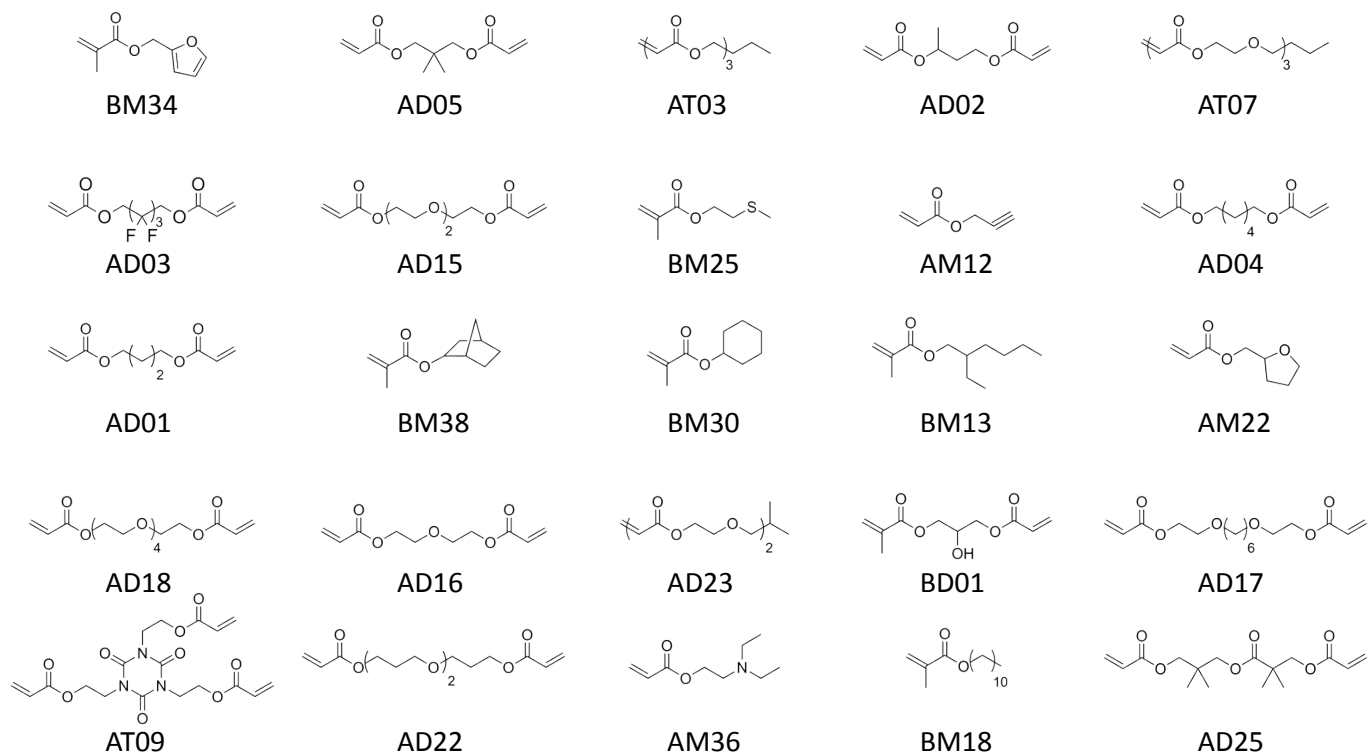
Supporting Figure S7. Monomer identities of top 25 materials.
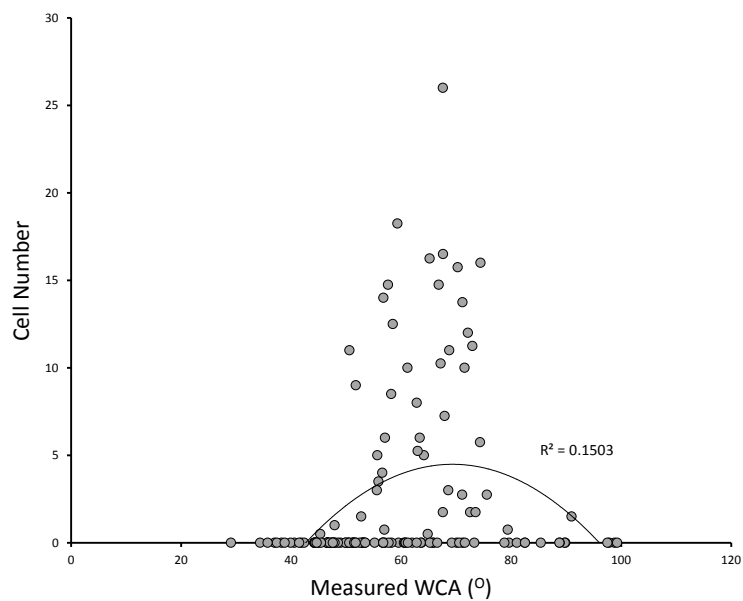
Supporting Figure S8. Measured cell number versus wettability for 141 homopolymers.
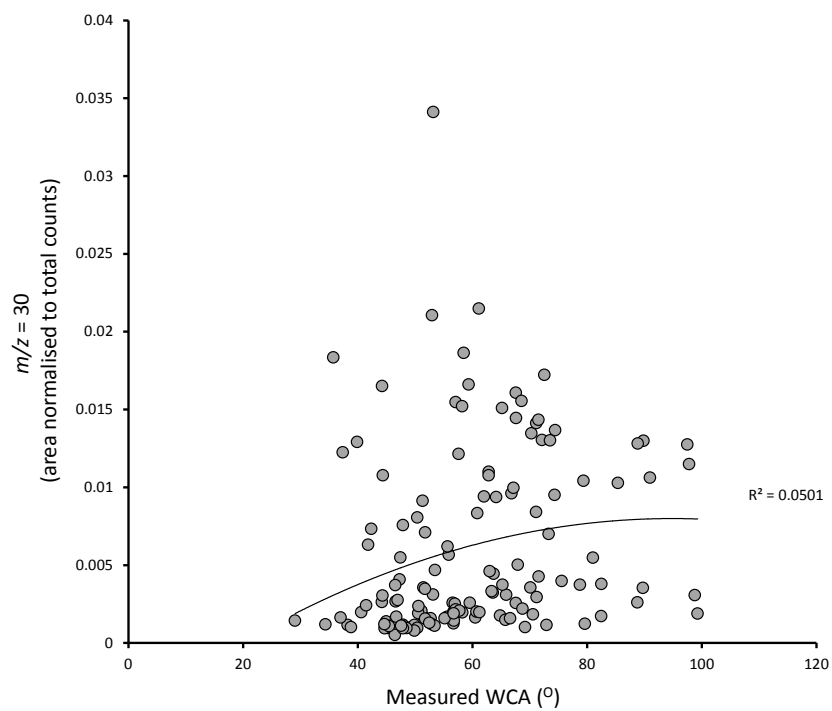
Supporting Figure S9. Normalised intensity in the *m/z* = 30 region versus wettability for 141 homopolymers.