Supplementary Materials

First Stage of the Development of an Eco-Friendly Detergent Formulation for Efficient Removal of Carbonized Soil

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Contents

Number of pages: 14

Number of figures: 6

Number of tables: 11

In this work, the commercial degreaser KH7, from KH Lloreda company, was used. This degreaser is composed of water, fatty ethoxylated alcohol, methoxypropanol, monoethanolamine and polycarboxylate with a pH of 11.25.

Tables

Table S1. pH of the different formulations used in the screening.

	pH				
Solvent → **Surfactant ↓**	PM	DPM	BDG	MMB	IPG
$[N_{1118}][C_8O_2]$	10.36	7.91	8.53	8.54	10.26
$[N_{1118}][C_{10}O_2]$	8.87	8.20	8.28	8.28	8.75
$[N_{1118}][C_{12}O_2]$	9.02	8.48	9.06	8.84	8.97
$[N_{11110}][C_8O_2]$	10.19	8.32	9.82	7.86	10.38
$[N_{11110}][C_{10}O_2]$	9.21	8.45	8.83	8.83	9.06
$[N_{11110}][C_{12}O_2]$	9.30	8.56	8.33	8.10	9.34
$[N_{11112}][C_{10}O_2]$	9.35	8.39	8.75	9.15	9.25
$[N_{11112}][C_{12}O_2]$	9.65	8.62	9.00	9.30	9.46
C_{12}-C_{15} 7EO's	5.08	4.15	4.99	4.05	5.11
C_{10} 6EO's	4.68	4.04	4.67	3.18	4.54
C_{12}-C_{15} 9EO's	5.80	4.30	5.94	4.19	5.43
C_{11}-C_{13} 9EO's	5.07	3.95	4.99	3.90	4.71
C_{10}-C_{14} 8EO's	5.10	4.05	4.86	3.98	5.04

Table S2. *HLB* values of nonionic surfactants, given by the suppliers.

Nonionic surfactant	*HLB*
C_{10} 6EO′s	12.4
C_{11}-C_{13} 9EO′s	13.2
C_{12}-C_{15} 7EO′s	12.3
C_{12}-C_{15} 9EO's	13.1
C_{10}-C_{14} 8EO's	13.6

Table S3. Mixture design for optimization of the solvent composition.

Run	Coded variables		
	Surfactant (wt%)	Solvent (wt%)	Water (wt%)
1	10.00	13.00	77.00
2	10.00	3.00	87.00
3	3.00	13.00	84.00
4	3.00	3.00	94.00
5	3.00	8.00	89.00
6	10.00	8.00	82.00
7	6.50	3.00	90.50
8	6.50	13.00	80.50
9	6.50	8.00	85.50

Table S4. pH and efficiency obtained for the formulations used in the mixture design of C_{11}-C_{13} 9EO's, IPG and water.

Model for ceramic: R^2 = 0.97 and $R^2_{adj.}$ = 0.91

Model for stainless-steel: R^2 = 0.80 and R^2_{adj}= 0.47

Run	pH	Experimental Efficiency		Predicted Efficiency	
		Ceramic	Stainless-steel	Ceramic	Stainless-steel
1	4.24	0.45	0.65	0.42	0.61
2	4.09	0.76	0.87	0.79	0.80
3	4.24	1.02	0.87	0.99	0.93
4	4.18	0.60	0.90	0.62	0.92
5	4.19	0.90	0.90	0.90	0.83
6	4.15	0.70	0.51	0.70	0.61
7	4.30	0.93	0.82	0.87	0.86
8	4.20	0.82	0.80	0.87	0.78
9	4.14	0.96	0.74	0.97	0.72

Table S5. pH and efficiency obtained for the formulations used in the mixture design of $[N_{1118}][C_8O_2]$, BDG and water.

Model for ceramic: $R^2 = 0.94$ and $R^2_{adj.} = 0.84$

Model for stainless-steel: $R^2 = 0.92$ and $R^2_{adj} = 0.80$

Run	pH	Experimental Efficiency		Predicted Efficiency	
		Ceramic	Stainless-steel	Ceramic	Stainless-steel
1	8.23	0.87	1.10	0.83	1.06
2	8.74	0.63	0.95	0.65	0.93
3	7.58	0.47	1.03	0.41	0.99
4	7.92	0.32	0.41	0.31	0.40
5	7.70	0.25	0.65	0.33	0.70
6	8.41	0.68	0.93	0.70	0.99
7	7.83	0.87	0.68	0.87	0.71
8	8.05	0.90	0.98	1.00	1.06
9	8.15	1.00	1.00	0.90	0.89

Table S6. ANOVA for the mixture design using formulations composed of C_{11}-C_{13} 9EOs, IPG and water applied to the ceramic surface.

	SS	df	MS	F	p
Model (Regression)	0.265929	5	0.053186	17.30503	0.020203
Total Error (Residual)	0.009220	3	0.003073		
Total Adjusted	0.275149	8	0.034394		

Table S7. ANOVA for the mixture design using formulations composed of C_{11}-C_{13} 9EOs, IPG and water applied to the stainless-steel surface.

	SS	df	MS	F	p
Model (Regression)	0.110659	5	0.022132	3.399777	0.051053
Total Error (Residual)	0.027667	3	0.009222		
Total Adjusted	0.138327	8	0.017291		

Table S8. Efficiency and pH obtained for the optimal composition of the formulation composed of C_{11}-C_{13} 9EO's, IPG and water.

Surface	Optimal composition			Efficiency		Relative deviation (%)	pH
	IPG	C_{11}-C_{13} 9EO's	H_2O	Experimental	Predicted		
Ceramic	10.0	5.0	85.0	1.03	1.00	3.00	4.30
Stainless-steel	13.0	3.0	84.0	0.95	0.91	4.40	4.24

Table S9. ANOVA for the mixture design using formulations composed of $[N_{1118}][C_8O_2]$, BDG and water applied to the ceramic surface.

	SS	df	MS	F	p
Model (Regression)	0.54402678	5	0.108805	9.313672	0.04783
Total Error (Residual)	0.03504698	3	0.011682		
Total Adjusted	0.57907375	8	0.072384		

Table S10. ANOVA for the mixture design using formulations composed of $[N_{1118}][C_8O_2]$, BDG and water applied to the stainless-steel surface.

	SS	df	MS	F	p
Model (Regression)	0.381089	5	0.076218	7.227412	0.047155
Total Error (Residual)	0.031637	3	0.010546		
Total Adjusted	0.412726	8	0.051591		

Table S11. Efficiency and pH obtained for the optimal composition of the formulation composed of $[N_{1118}][C_8O_2]$, BDG and water.

Surface	Optimal composition			Efficiency		Relative deviation (%)	pH
	BDG	$[N_{1118}][C_8O_2]$	H_2O	Experimental	Predicted		
Ceramic	13.0	7.0	80.0	0.99	1.03	3.90	8.28
Stainless-steel	13.0	8.0	79.0	1.04	1.07	2.80	8.63

Figures

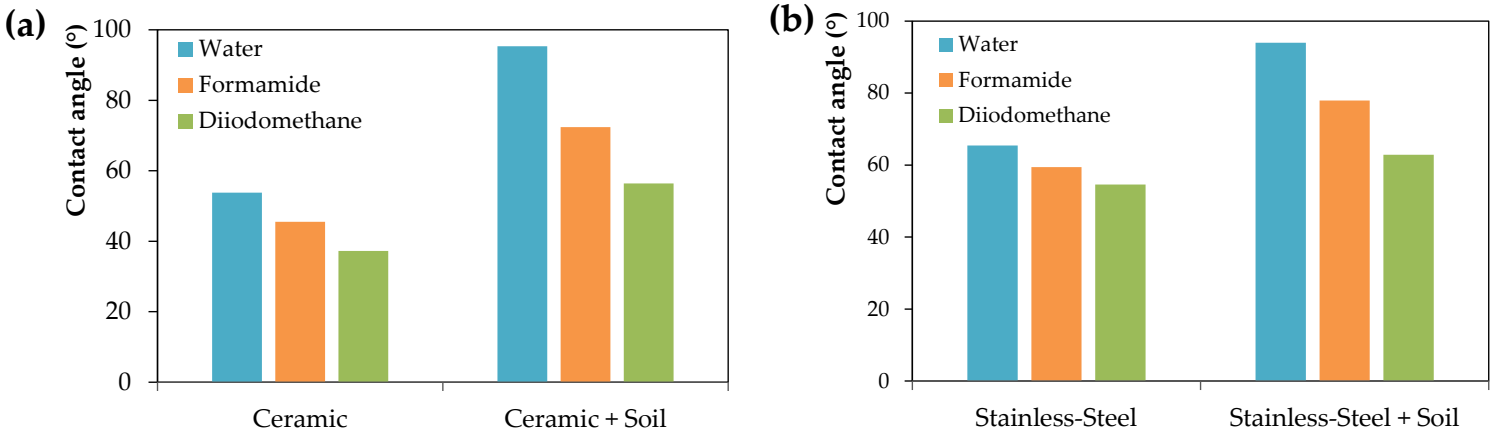


Figure S1. Contact angles of net and soiled surfaces: (a) ceramic and (b) stainless-steel.

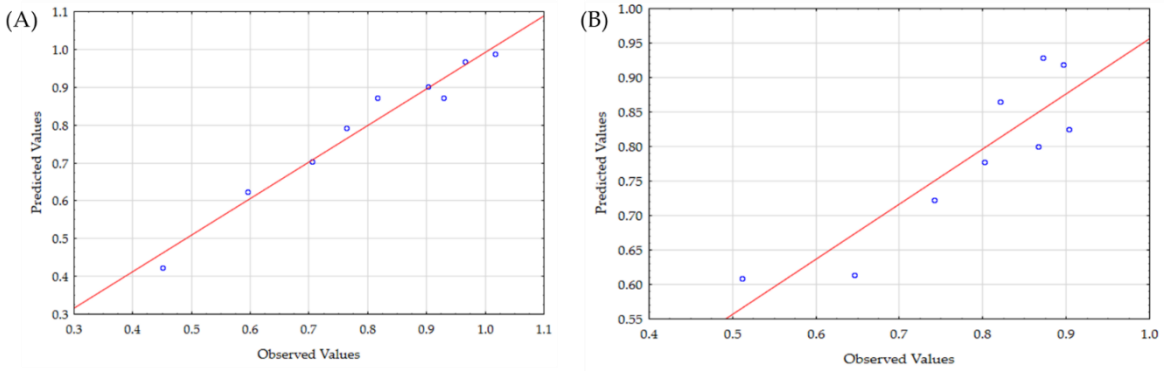


Figure S2. Predict *vs.* observed values of IPG + C_{11}-C_{13} 9EO's for soil's removal from (A) ceramic and (B) stainless-steel.

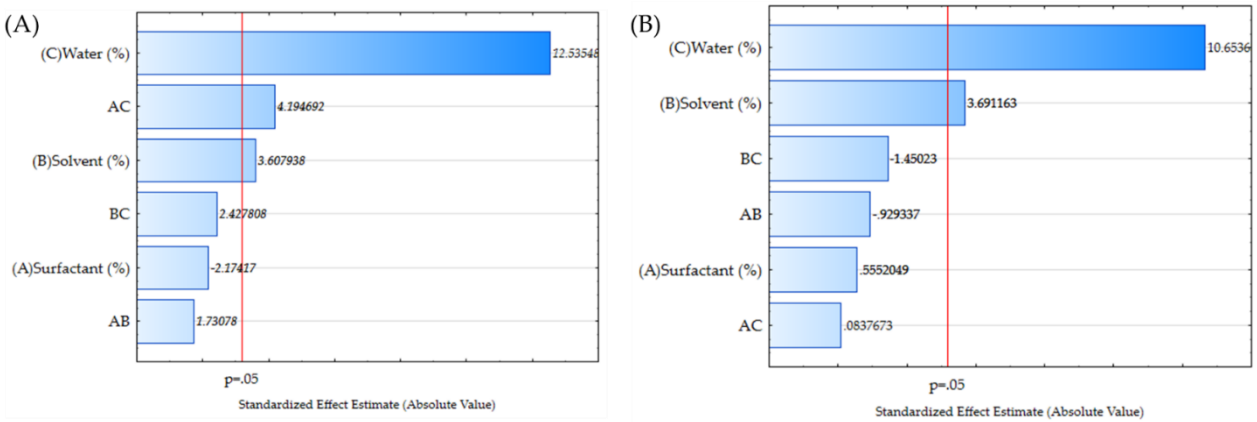


Figure S3. Pareto charts for the standardized main effects in the IPG + C_{11}-C_{13} 9EO's mixture design for (A) ceramic and (B) stainless-steel. The vertical line indicates the statistical significance of the effects (95% of confidence).

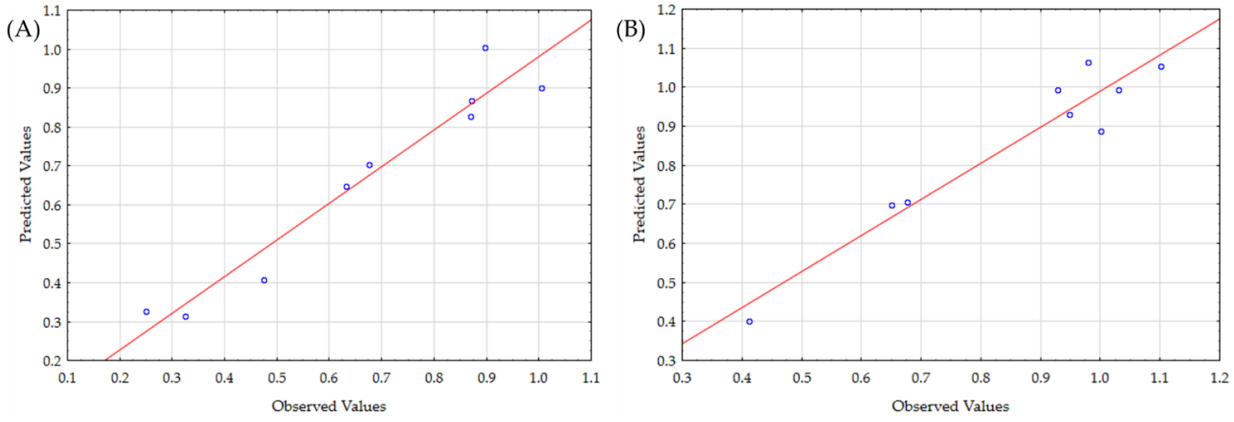


Figure S4. Predict *vs.* observed values of BDG + $[N_{1118}][C_8O_2]$ for soil's removal from (A) ceramic and (B) stainless-steel.

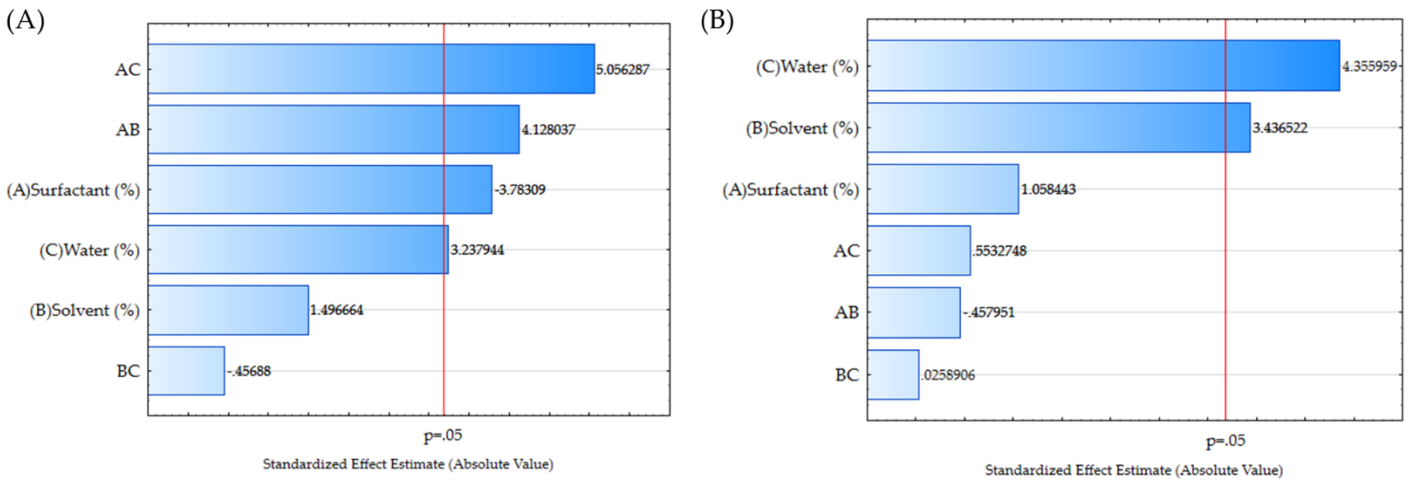
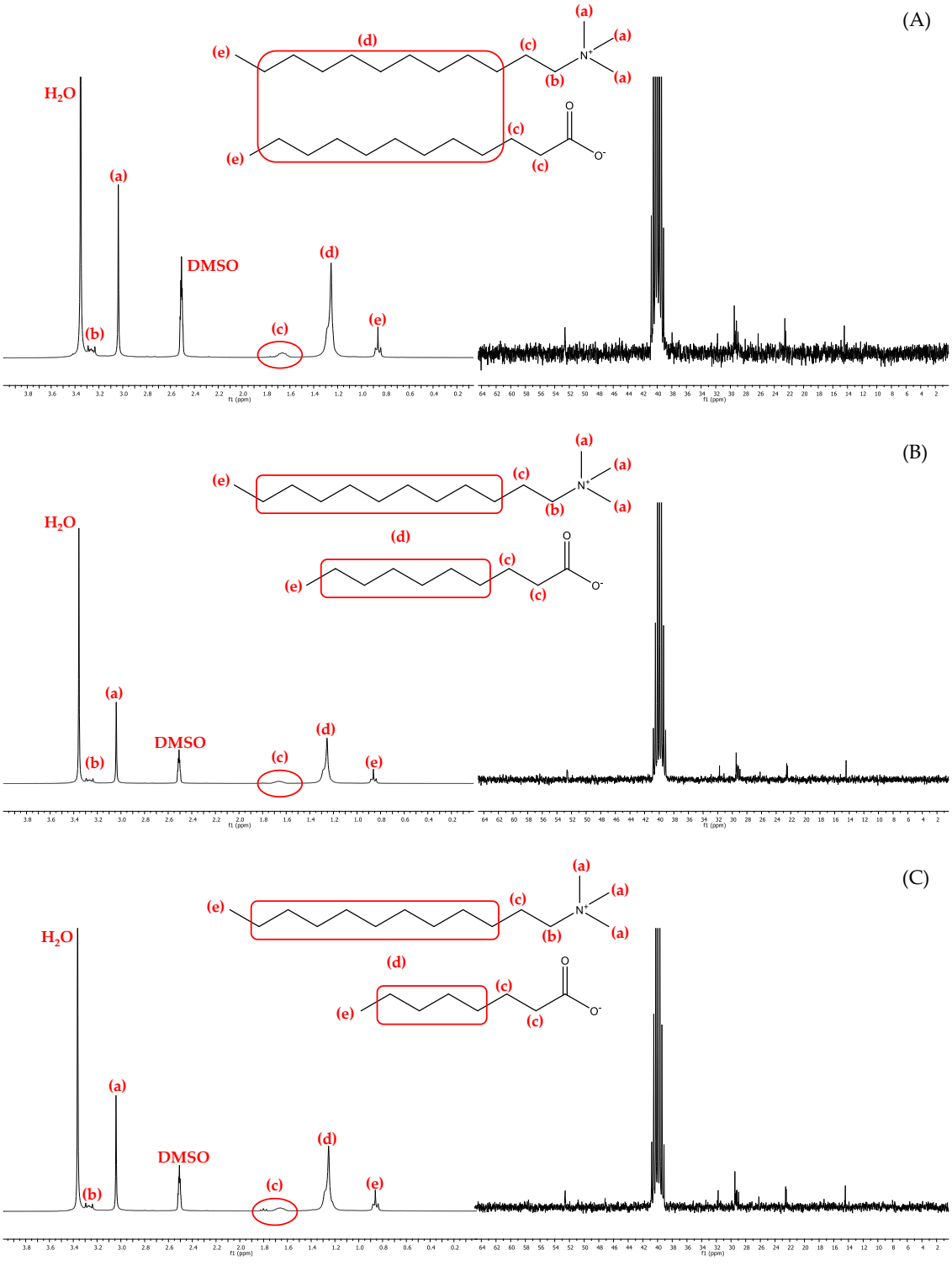
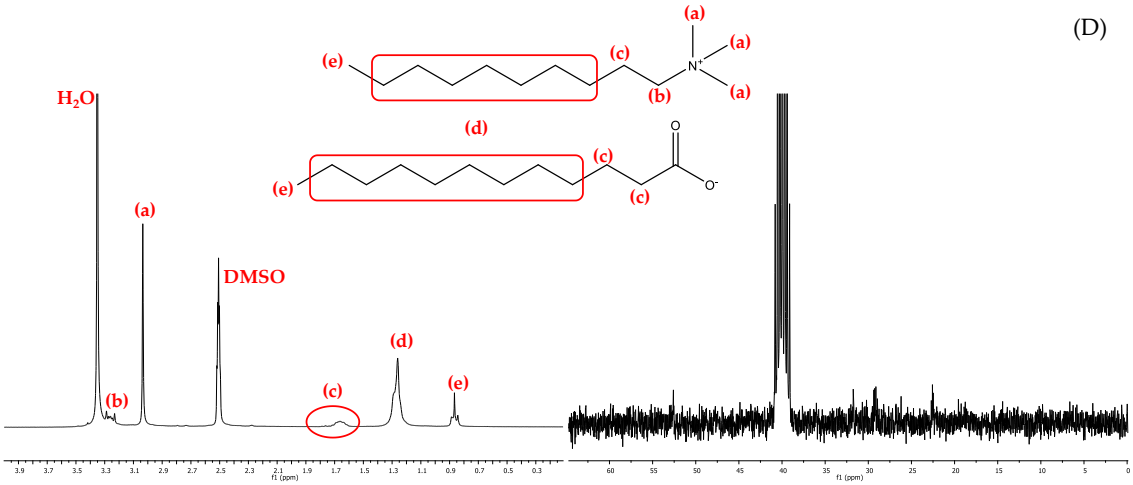
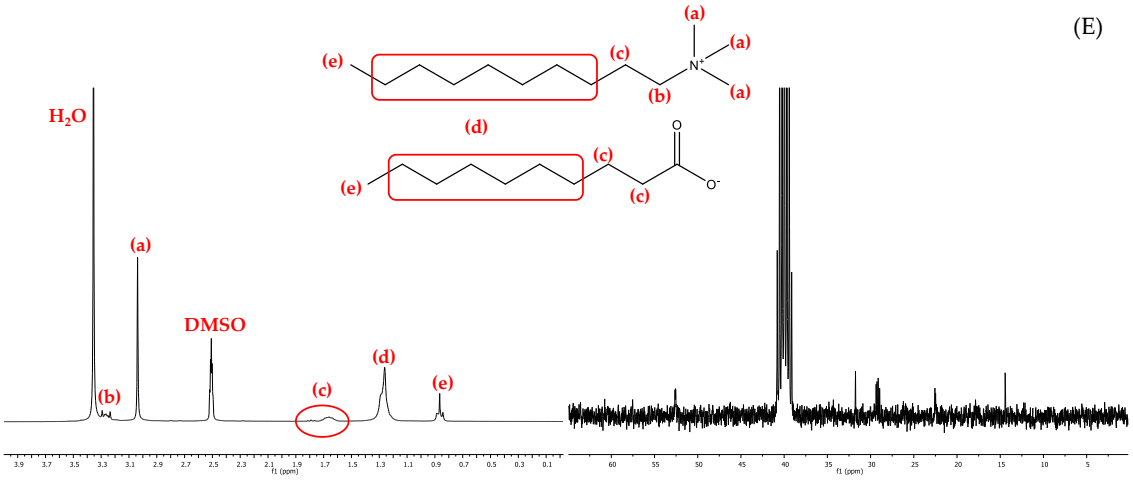
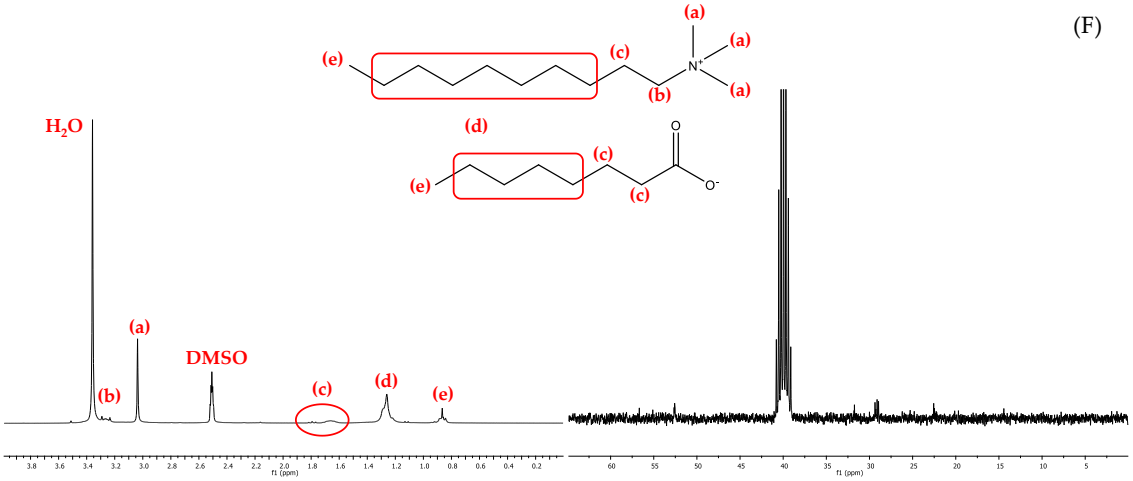


Figure S5. Pareto charts for the standardized main effects in the BDG + $[N_{1118}][C_8O_2]$ mixture design for (A) ceramic and (B) stainless-steel. The vertical line indicates the statistical significance of the effects (95% of confidence).


(A)
H2O
(a)
DMSO
(b)
(c)
(d)
(e)
(B)
H2O
(a)
DMSO
(b)
(c)
(d)
(e)
(C)
H2O
(a)
DMSO
(b)
(c)
(d)
(e)


(D)
H2O
(a)
DMSO
(b)
(c)
(d)
(e)
N+
O-


(E)
H2O
(a)
DMSO
(b)
(c)
(d)
(e)
N+
O-


(F)
H2O
(a)
DMSO
(b)
(c)
(d)
(e)
N+
O-

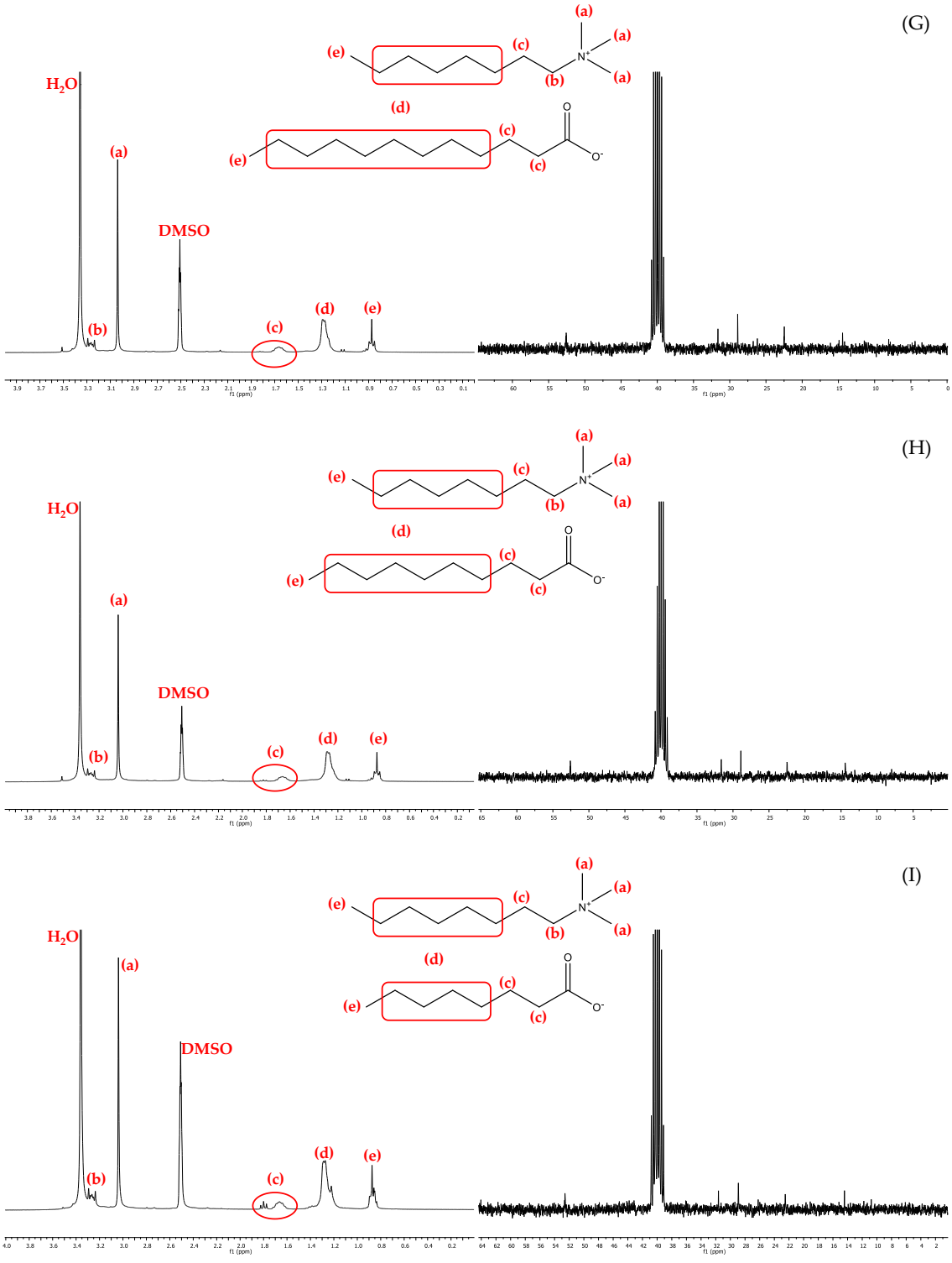


Figure S6. ^{1}H (left panel) and ^{13}C (right panel) NMR spectra in DMSO of the ionic surfactants synthesized: (A) $[N_{11112}][C_{12}O_2]$; (B) $[N_{11112}][C_{10}O_2]$; (C) $[N_{11112}][C_8O_2]$; (D) $[N_{11110}][C_{12}O_2]$; (E) $[N_{11110}][C_{10}O_2]$; (F) $[N_{11110}][C_8O_2]$; (G) $[N_{1118}][C_{12}O_2]$; (H) $[N_{1118}][C_{10}O_2]$; (I) $[N_{1118}][C_8O_2]$.