Supporting Information

Radiation Effect of Carboxyl-Functionalized Task-Specific Ionic Liquids on UO_2^{2+} Removal: Experimental Study with DFT Validation

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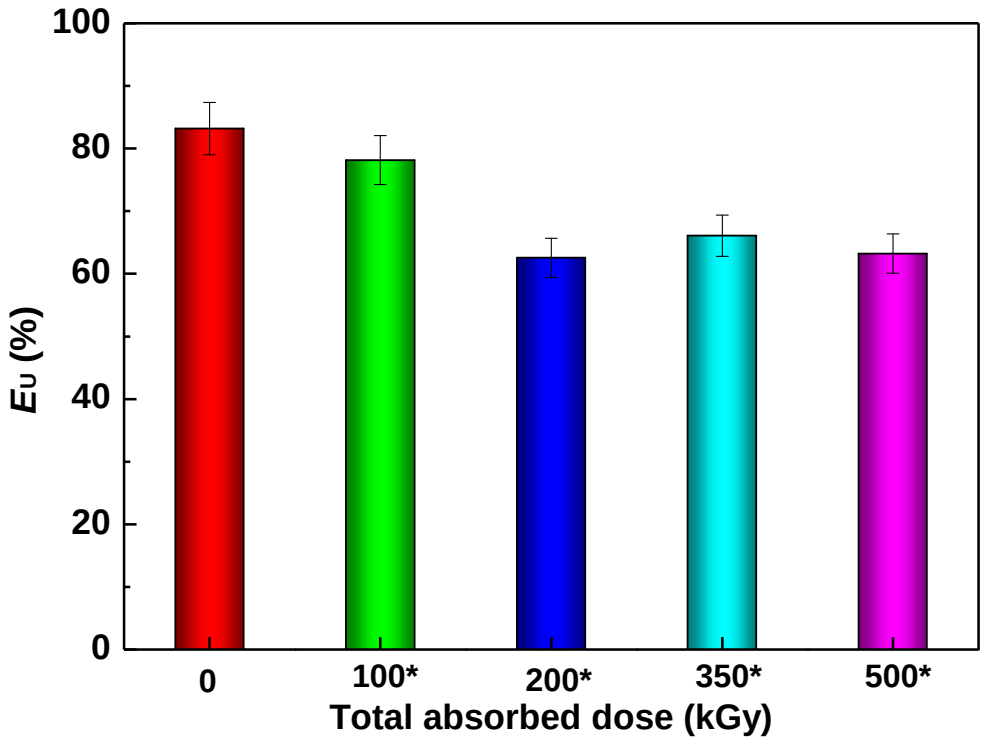


Figure S1. Influence of dose on UO_2^{2+} extraction by water-washed [$HOOCCH_2MIM$][NTf_2].

*These irradiated samples were washed for 4 times by deionized water before extraction.

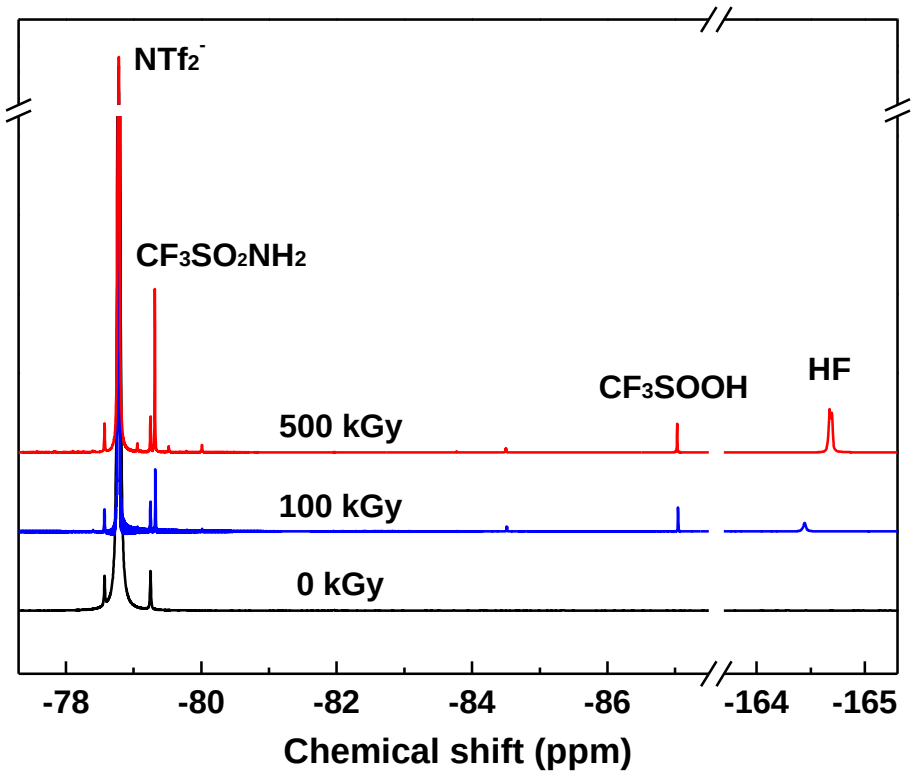


Figure S2. ^{19}F NMR spectra of [$HOOCCH_2MIM$][NTf_2] before and after irradiation.

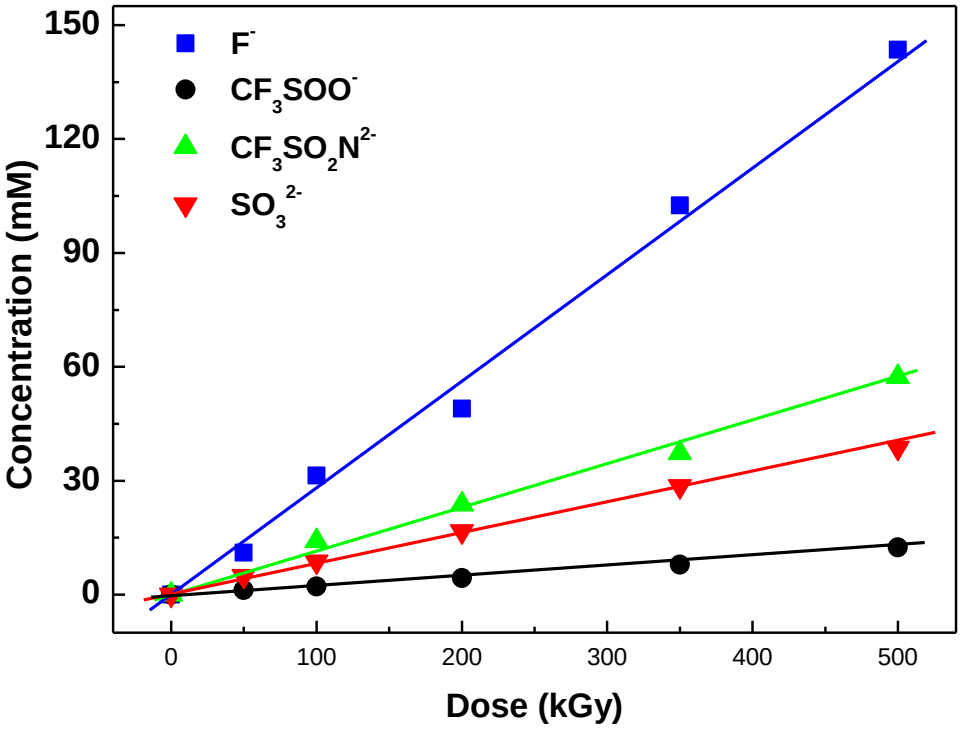


Figure S3. The relationship between the concentration of water-soluble radiolytic products and radiation doses.

Table S1. Designation of signals of [$HOOCCH_2MIM$][NTf_2] before and after γ-irradiation.

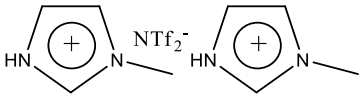
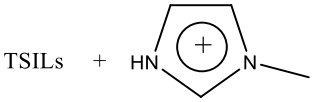
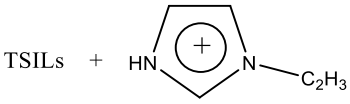
Experimental m/z	**Designation**	**Theoretical m/z**
281.126	[[$HOOCCH_2MIM$]+[$^-OOCCH_2MIM$]]$^+$	281.124
446.037		446.039
504.044		504.044
516.043		516.044
562.048	[[$HOOCCH_2MIM$][NTf_2]+[$HOOCCH_2MIM$]]$^+$	562.050
702.105	[[$HOOCCH_2MIM$]$_2$[NTf_2]+[$^-OOCCH_2MIM$]]$^+$	702.108
842.162	[[$HOOCCH_2MIM$]$_2$[NTf_2]$_2$+[$^-OOCCH_2MIM$]$_2$]$^+$	842.167
1123.084	[[$HOOCCH_2MIM$]$_3$[NTf_2]$_2$+[$^-OOCCH_2MIM$]]$^+$	1123.092
1263.141	[[$HOOCCH_2MIM$]$_3$[NTf_2]$_2$+[$^-OOCCH_2MIM$]$_2$]$^+$	1263.150
1404.009	[[$HOOCCH_2MIM$]$_4$[NTf_2]$_3$]$^+$	1404.017

Table S2. The enthalpy, entropy, and binding energies (298.15 K) for radiolytic products, metal ions and complexes obtained in the gas phase by DFT method at B3LYP/6-31+G(d,p)/RECP level.

Species	H_g (a.u.)	TS_g (kJ/mol)	G_g (a.u.)
[OOCCH$_2$MIM]	-493.252	123.30	-493.299
UO_2^{2+}	-626.744	79.54	-626.774
$[UO_2([OOCCH_2MIM])_3]^{2+}$	-2107.116	306.19	-2107.233
SO_3^{2-}	-623.739	82.51	-623.770
UO_2SO_3	-1251.437	114.77	-1251.481
CF_3SOO^-	-886.291	106.02	-886.331
$[UO_2CF_3SOO]^+$	-1513.550	136.11	-1513.602
$CF_3SO_2NH^-$	-941.598	108.66	-941.639
$[UO_2CF_3SO_2NH]^+$	-1568.851	141.92	-1568.905
$UO_2(CF_3SOO)_2$	-2400.061	196.90	-2400.136
$UO_2(CF_3SO_2NH)_2$	-2510.653	201.67	-2510.730
H_2O	-76.409	57.98	-76.431
$[UO_2(H_2O)_5]^{2+}$	-1009.169	169.99	-1009.234
F^-	-99.857	43.39	-99.874
UO_2F_2	-827.382	157.90	-827.442
$[UO_2F_4]^{2-}$	-1027.230	121.46	-1027.276
$[UO_2F_5]^{3-}$	-1126.855	133.13	-1126.906

Basis set: 6-31+G(d,p) for C, H, O, N, F, S; ECP60MWB for U(VI).

Table S3. The enthalpy, entropy, and binding energies (298.15 K) for radiolytic products, metal ions and complexes obtained in the water by DFT method at M05-2X/6-31G(d)/RECP level.

Species	H_{aq} (a.u.)	TS_{aq} (kJ/mol)	G_{aq} (a.u.)
[OOCCH$_2$MIM]	-493.209	120.49	-493.255
UO_2^{2+}	-627.169	79.91	-627.200
$[UO_2([OOCCH_2MIM])_3]^{2+}$	-2106.939	274.30	-2107.044
SO_3^{2-}	-623.998	81.93	-624.029
UO_2SO_3	-1251.287	112.85	-1251.330
CF_3SOO^-	-886.258	102.75	-886.297
$[UO_2CF_3SOO]^+$	-1513.469	134.11	-1513.520
$CF_3SO_2NH^-$	-941.561	107.01	-941.601
$[UO_2CF_3SO_2NH]^+$	-1568.755	139.52	-1568.808
$UO_2(CF_3SOO)_2$	-2399.766	188.40	-2399.838
$UO_2(CF_3SO_2NH)_2$	-2510.338	196.48	-2510.413
H_2O	-76.383	58.00	-76.405
$[UO_2(H_2O)_5]^{2+}$	-1009.205	155.97	-1009.264
F^-	-99.896	43.37	-99.912
UO_2F_2	-827.226	103.53	-827.265
$[UO_2F_4]^{2-}$	-1027.237	125.57	-1027.285
$[UO_2F_5]^{3-}$	-1127.179	123.40	-1127.226

Basis set: 6-31G(d) for C, H, O, N, F, S; ECP60MWB for U(VI).

Table S4. The enthalpy, entropy, and binding energies (298.15 K) for $[UO_2F_n]^{2-n}$ (n = 2, 4, 5) in the organic and the aqueous phase by DFT method at M05-2X/6-31G(d)/RECP level.

Species	H_{org} (a.u.)	TS_{org} (kJ/mol)	G_{org} (a.u.)	H_{aq} (a.u.)	TS_{aq} (kJ/mol)	G_{aq} (a.u.)
UO_2^{2+}	-627.089	70.31	-627.116	-627.169	79.91	-627.200
H_2O	-627.089	70.31	-627.116	-76.383	58.00	-76.405
$[UO_2(H_2O)_5]^{2+}$	-1009.136	160.91	-1009.197	-1009.205	155.97	-1009.264
F^-	-99.898	43.37	-99.914	-99.896	43.37	-99.912
UO_2F_2	-827.200	100.40	-827.238	-827.226	103.53	-827.265
$[UO_2F_4]^{2-}$	-1027.227	120.70	-1027.272	-1027.237	125.57	-1027.285
$[UO_2F_5]^{3-}$	-1127.179	123.40	-1127.226	-1127.201	125.01	-1127.249

Basis set: 6-31G(d) for O, F; ECP60MWB for U(VI).