

## Supplementary information

Table S1: EXAFS fit results. N is the coordination number ( $\pm 20\%$ ), R is the interatomic distance ( $\pm 0.01 \text{ \AA}$ ),  $\sigma^2$  is the Debye-Waller factor ( $\pm 0.001 \text{ \AA}^2$ ),  $E_0$  is the shift in energy.  $R_{\text{factor}}$  indicates the goodness of the fit according to <sup>37</sup>.

\* Fixed parameter, # linked parameter

Sample ID	paths	N	R ( $\text{\AA}$ )	$\sigma^2$ ( $\text{\AA}^2$ )	$E_0$ (eV)	$R_{\text{factor}}$
Sample A	Oax	2*	1.78	0.002	5.1	0.04
	O (DMDBMA)	5.8	2.38	0.007		
	C	4.8	3.42	0.006		
	MS ax	2#	3.56#	0.004*		
	MS C-O-C	11.6#	3.53#	0.008#		
	MS O-C-O-C	5.8#	3.60#	0.008#		
S1_0_1	Oax	2*	1.77	0.002	4.5	0.06
	O	6.0	2.40	0.010		
	C	2.0	3.45	0.007		
	MS ax	2#	3.54#	0.004*		
	MS C-O-C	4#	3.55#	0.008#		
	MS O-C-O-C	2#	3.63#	0.008#		
S1_0_2	Oax	2*	1.77	0.002	4.5	0.05
	O (DMDBMA)	5.7	2.38	0.008		
	C	4.0	3.43	0.005		
	MS ax	2#	3.55#	0.004#		
	MS C-O-C	8#	3.53#	0.008#		
	MS O-C-O-C	4#	3.61#	0.008#		
S1_0_3	Oax	2*	1.78	0.002	4.5	0.05
	O (DMDBMA)	5.7	2.37	0.007		
	C	4.5	3.42	0.005		
	MS ax	2#	3.56#	0.004#		
	MS C-O-C	9#	3.53#	0.008#		
	MS O-C-O-C	4.5#	3.61#	0.008#		
S1_0_4	Oax	2*	1.77	0.002	3.2	0.05
	O (DMDBMA)	5.7	2.37	0.007		
	C	5.8	3.42	0.006		
	MS ax	2#	3.56#	0.004#		
	MS C-O-C	11.4#	3.53#	0.008#		
	MS O-C-O-C	5.7#	3.60#	0.008#		
Sample B	Oax	2*	1.77	0.002	5.0	0.03
	O (DMDBMA)	2.2	2.37	0.005		
	O ( $\text{NO}_3^-$ )	3.8	2.52	0.005		
	C	2.2#	3.47	0.004		
	MS ax	2#	3.55#	0.003#		
	MS C-O-C	4.5#	3.55#	0.008#		
	MS O-C-O-C	2.2#	3.62#	0.007#		
	N	1.9	2.95	0.003		
	Od	1.9#	4.21	0.005		
	MS Od	3.8#	4.21#	0.005#		
	MS Od	1.9#	4.21#	0.005#		
S1_1_2	O	2*	1.78	0.002	5.2	0.03
	O (DMDBMA)	3.6	2.36	0.005		
	O ( $\text{NO}_3^-$ )	2.4	2.51	0.005		
	C	3.6#	3.44	0.006		
	MS ax	2#	3.56#	0.003#		
	MS C-O-C	7.3#	3.53#	0.008#		
	MS O-C-O-C	3.6#	3.61#	0.007#		
	N	1.2#	2.93	0.004		
	Od	1.2#	4.21	0.006		
	MS Od	2.4#	4.21#	0.006#		
	MS Od	1.2#	4.21#	0.006#		

S1_2_2	O	2*	1.78	0.002	5.3	0.05
	O(DMDBMA)	3.0	2.36	0.006		
	O (NO <sub>3</sub> <sup>-</sup> )	3.0	2.52	0.006		
	C	3.0 <sup>#</sup>	3.45	0.005		
	MS ax	2 <sup>#</sup>	3.56 <sup>#</sup>	0.004 <sup>#</sup>		
	MS C-O-C	6.0 <sup>#</sup>	3.54 <sup>#</sup>	0.008 <sup>#</sup>		
	MS O-C-O-C	3.0 <sup>#</sup>	3.61 <sup>#</sup>	0.007 <sup>#</sup>		
	N	1.5 <sup>#</sup>	2.95	0.003		
	Od	1.5 <sup>#</sup>	4.21	0.005		
	MS Od	3.0 <sup>#</sup>	4.21 <sup>#</sup>	0.005 <sup>#</sup>		
MS Od	1.5 <sup>#</sup>	4.21 <sup>#</sup>	0.005 <sup>#</sup>			
S1_3_2	O	2*	1.78	0.002	4.5	0.04
	O(DMDBMA)	2.6	2.37	0.006		
	O (NO <sub>3</sub> <sup>-</sup> )	3.4	2.52	0.006		
	C	2.6 <sup>#</sup>	3.45	0.006		
	MS ax	2 <sup>#</sup>	3.56 <sup>#</sup>	0.003 <sup>#</sup>		
	MS C-O-C	5.1 <sup>#</sup>	3.54 <sup>#</sup>	0.008 <sup>#</sup>		
	MS O-C-O-C	2.6 <sup>#</sup>	3.62 <sup>#</sup>	0.008 <sup>#</sup>		
	N	1.7 <sup>#</sup>	2.95	0.004		
	Od	1.7 <sup>#</sup>	4.21	0.005		
	MS Od	3.4 <sup>#</sup>	4.21 <sup>#</sup>	0.005 <sup>#</sup>		
MS Od	1.7 <sup>#</sup>	4.21 <sup>#</sup>	0.005 <sup>#</sup>			
S1_2_1	O	2*	1.78	0.002	5.5	0.05
	O(DMDBMA)	2.0	2.36	0.006		
	O (NO <sub>3</sub> <sup>-</sup> )	4.0	2.51	0.006		
	C	2.0 <sup>#</sup>	3.46	0.005		
	MS ax	2 <sup>#</sup>	3.56 <sup>#</sup>	0.003 <sup>#</sup>		
	MS C-O-C	4.0 <sup>#</sup>	3.54 <sup>#</sup>	0.008 <sup>#</sup>		
	MS O-C-O-C	2.0 <sup>#</sup>	3.61 <sup>#</sup>	0.008 <sup>#</sup>		
	N	2.0 <sup>#</sup>	2.95	0.003		
	Od	2.0 <sup>#</sup>	4.21	0.005		
	MS Od	4.0 <sup>#</sup>	4.21 <sup>#</sup>	0.005 <sup>#</sup>		
MS Od	2.0 <sup>#</sup>	4.21 <sup>#</sup>	0.005 <sup>#</sup>			
DCE	O	2*	1.77	0.002	4.6	0.03
	O(DMDBMA)	2.3	2.36	0.004		
	O (NO <sub>3</sub> <sup>-</sup> )	3.7	2.51	0.004		
	C	2.3 <sup>#</sup>	3.45	0.005		
	MS ax	2 <sup>#</sup>	3.56 <sup>#</sup>	0.003 <sup>#</sup>		
	MS C-O-C	4.6 <sup>#</sup>	3.53 <sup>#</sup>	0.008 <sup>#</sup>		
	MS O-C-O-C	2.3 <sup>#</sup>	3.61 <sup>#</sup>	0.011 <sup>#</sup>		
	N	1.8 <sup>#</sup>	2.94	0.002		
	Od	1.8 <sup>#</sup>	4.20	0.006		
	MS Od	3.7 <sup>#</sup>	4.20 <sup>#</sup>	0.006 <sup>#</sup>		
MS Od	1.8 <sup>#</sup>	4.20 <sup>#</sup>	0.006 <sup>#</sup>			

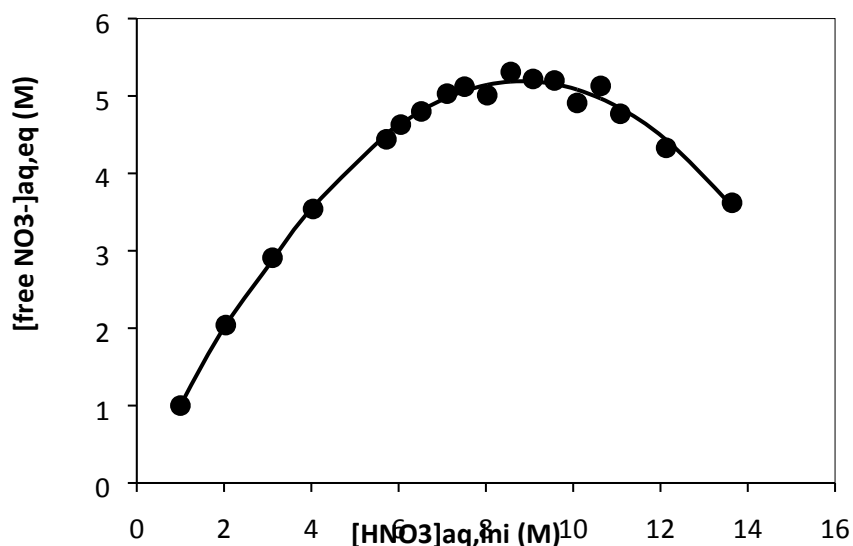


Figure S1: Free nitrate concentration in aqueous solution as a function of the initial nitric acid concentration, as given by <sup>44</sup> (dots). The solid line represents the data fit used in this study.

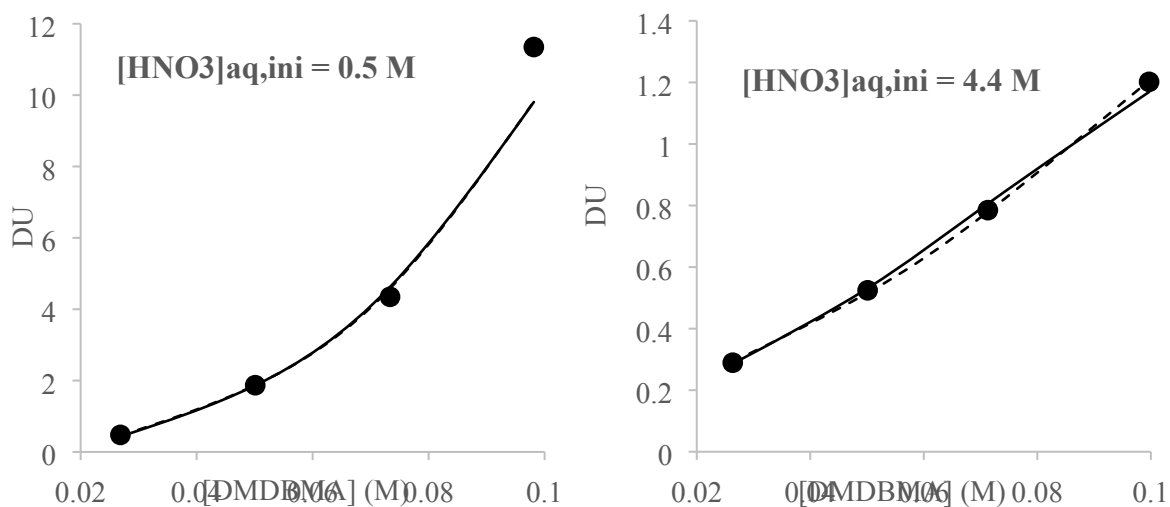


Figure S2: Evolution of the uranium distribution coefficient as a function of the DMBMA concentration in the IL phase, at two HNO<sub>3</sub> concentrations: 0.5 M (left) and 4.4 M (right). Lines represent data fits obtained by the model explained in part 4, taking into account the aqueous Tf<sub>2</sub>N<sup>-</sup> concentrations values: model from <sup>34</sup> (dotted lines) and new model developed in this paper (solid lines).