

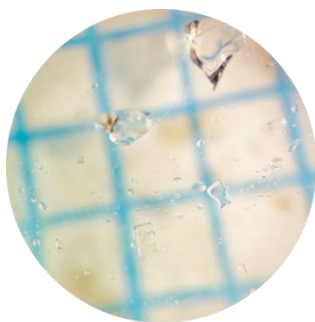
Table S1. Geometry of hydrogen bonds (Å, °) for compounds **H₂EtGlyCl**, **H₂(*i*-PrGly)Cl**, **H₂(*n*-PrGly)Cl**, **H₂EtGlyNO₃**, **H₂(*i*-PrGly)NO₃**, **H₂(*n*-PrGly)NO₃** and **H(*n*-PrGly)·1/3H₂O**

	D–H...A	D–H (Å)	H...A (Å)	D...A (Å)	D–H...A(°)
H₂EtGlyCl	N1–H1A...Cl1 ^a	0.868(16)	2.311(16)	3.1638(8)	167.5(14)
	O2–H2...Cl1 ^b	0.79(2)	2.28(2)	3.0593(14)	166.2(19)
H₂(<i>i</i>-PrGly)Cl	N1–H1A...Cl2 ^c	0.89	2.36	3.1840(14)	154
	N1–H1B...Cl1 ^d	0.89	2.26	3.1392(14)	171
	N2–H2A...Cl1 ^e	0.89	2.26	3.1352(15)	168
	N2–H2B...Cl2 ^e	0.89	2.34	3.1742(16)	157
	O12–H11...Cl1	0.82	2.25	3.0625(13)	170
	O22–H21...Cl2 ^f	0.82	2.19	2.9783(13)	163
H₂(<i>n</i>-PrGly)Cl	N1–H1N...Cl1 ^g	0.880(17)	2.290(17)	3.1475(8)	164.9(15)
	O2–H1...Cl1 ^h	0.90(3)	2.13(3)	3.0133(14)	166(3)
H₂EtGlyNO₃	N1–H1N...O2N ⁱ	0.85(3)	2.03(3)	2.866(2)	170(3)
	O2–H1...O2N ^a	0.82	2.2	2.937(3)	151
	O2–H1...O2N ^j	0.82	2.2	2.937(3)	151
H₂(<i>i</i>-PrGly)NO₃	N1–H1A...O21 ^k	0.89(3)	1.96(3)	2.855(2)	174(2)
	N1–H1B...O22 ^l	0.96(3)	1.92(3)	2.866(2)	171(2)
	N1–H1B...O23 ^l	0.96(3)	2.53(3)	3.165(3)	124(2)
	O12–H1...O21	0.92(4)	1.78(4)	2.671(2)	163(4)
	O12–H1...O22	0.92(4)	2.42(4)	3.136(2)	135(3)
H₂(<i>n</i>-PrGly)NO₃	N1–H1A...O21N ^m	0.89	2.1	2.895(3)	148
	N1–H1A...O23N ^m	0.89	2.4	3.226(3)	154
	N1–H1A...O11N	0.89	2.22	3.027(3)	151
	N1–H1A...O12N	0.89	2.35	3.103(3)	142
	N1–H1A...O22N	0.89	2.03	2.868(3)	157
	N1–H1A...O11N	0.89	2.26	2.960(3)	135
	N1–H1A...O13N	0.89	2.22	3.084(3)	162
	O12–H11...O12N ⁿ	0.82	2.58	3.099(3)	123
	O12–H11...O13N ⁿ	0.82	1.96	2.754(3)	163
	O22–H21...O22N ^o	0.82	2.04	2.774(3)	149
	O22–H21...O23N ^o	0.82	2.4	3.135(4)	150
H(<i>n</i>-PrGly)·1/3H₂O	N1–H1A...O1W ^p	0.91(3)	2.00(3)	2.828(3)	151(3)
	N1–H1B...O12 ^p	0.91(3)	1.93(3)	2.823(3)	168(2)
	N2–H2A...O32 ^r	0.81(2)	1.99(3)	2.795(3)	174(3)
	N2–H2B...O22 ^s	0.90(3)	1.84(3)	2.728(3)	168(3)
	N3–H3A...O21 ^p	0.87(3)	1.96(3)	2.776(3)	155(2)
	N3–H2B...O12 ^h	0.96(3)	1.87(3)	2.767(3)	154(3)
	O1W–H1W...O31 ^p	0.90(4)	1.90(4)	2.789(3)	171(3)
	O1W–H2W...O31 ^r	0.82(3)	1.98(3)	2.800(3)	176(3)

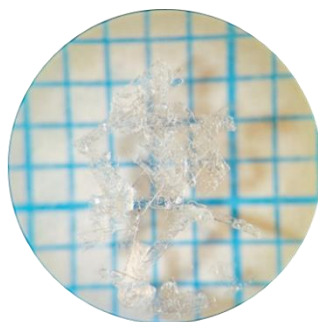
^a1/2-x, 1-y, -1/2+z, ^b-1/2+x, 1/2-y, 3/2-z, ^c1-x, 1-y, -z, ^d-x, 1-y, -z, ^e1-x, 2-y, -z, ^fx, 1+y, z, ^g-x, -1/2+y, 1-z, ^hx, y, 1+z, ⁱ-x, y, z, ^j1/2-x, 1-y, 1/2+z, ^k1-x, 1-y, -z, ^l1-x, -1/2+y, 1/2-z, ^mx, -1+y, z, ⁿ-x, -y, -z, ^o-x, 1-y, 1-z, ^p1-x, 1-y, 1/2+z, ^r1-x, 1-y, 1/2+z, ^s1-x, -y, -1/2+z

Table S2. Fractions of atomic contacts on a Hirshfeld surface of symmetrically independent *N*-alkylglycinium cations and zwitterions.

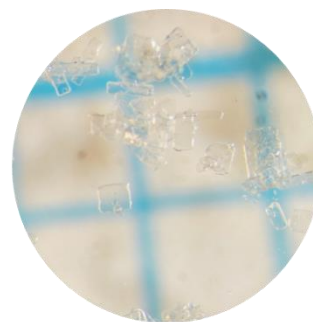
	O...O contacts / %	O...N contacts / %	O...H contacts / %	O...C contacts / %	N...N contacts / %	N...H contacts / %	N...C contacts / %	H...H contacts / %	H...C contacts / %	C...C contacts / %	O...Cl contacts / %	N...Cl contacts / %	H...Cl contacts / %	C...Cl contacts / %	Cl...Cl contacts / %
H₂EtGlyNO₃	3.3	0.8	64.4	0.4	0	1.4	0	27.9	1.7	0	/	/	/	/	/
H₂(<i>i</i>-PrGly)NO₃	3.7	0.4	55.5	2	0	1.5	0	36.1	0.7	0	/	/	/	/	/
H₂(<i>n</i>-PrGly)NO₃ – cation 1	3	0.9	56	0.9	0	1.3	0	36.7	1.3	0	/	/	/	/	/
H₂(<i>n</i>-PrGly)NO₃ – cation 2	2.4	0.8	53.5	0.5	0	0.9	0	40.4	1.4	0	/	/	/	/	/
H(<i>n</i>-PrGly)·1/3H₂O – zwitterion 1	0	0	40.7	0	0	0	0	57.4	1.8	0	/	/	/	/	/
H(<i>n</i>-PrGly)·1/3H₂O – zwitterion 2	0.1	0	40.5	0	0	0	0	55.2	4.2	0	/	/	/	/	/
H(<i>n</i>-PrGly)·1/3H₂O – zwitterion 3	0	0	41.5	0	0	0	0	54.8	3.6	0	/	/	/	/	/
H₂EtGlyCl	3.1	0	26.2	3.7	0	0	0	46.8	0.1	0	0	0	20	0	0
H₂(<i>i</i>-PrGly)Cl – cation 1	2.2	0	25.2	3.1	0	0	0	52.4	0.3	0	0.2	0	16.7	0	0
H₂(<i>i</i>-PrGly)Cl – cation 2	2.2	0	25.1	3	0	0	0	51.2	0.3	0	0	0	18.1	0	0
H₂(<i>n</i>-PrGly)Cl	1.6	0	24.7	2.9	0	0	0	52.4	0.4	0	0	0	18	0	0



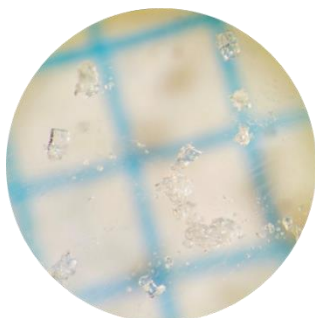
H₂EtGlyCl



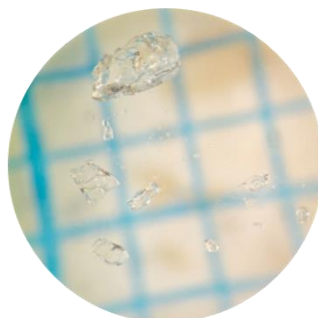
H₂(*i*-PrGly)Cl



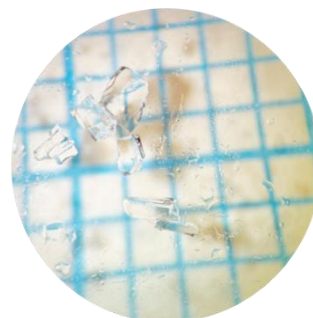
H₂(*n*-PrGly)Cl



H₂EtGlyNO₃



H₂(*i*-PrGly)NO₃

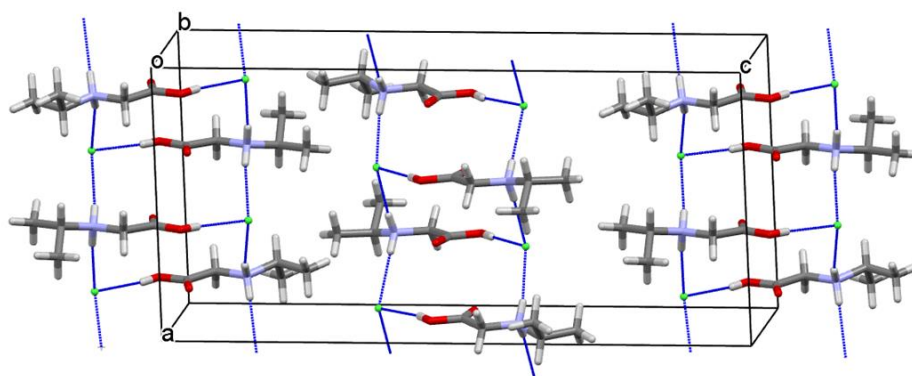


H₂(*n*-PrGly)NO₃

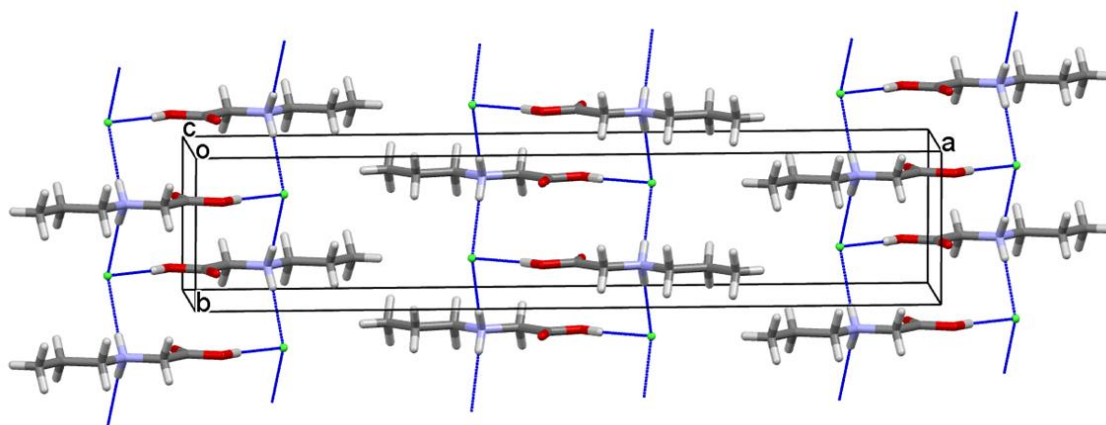


H(*n*-PrGly)NO₃·1/3H₂O

Figure S1. Microscopic images of the crystals of the *N*-alkylglycinium salts and the zwitterionic H(*n*-PrGly)·1/3H₂O.



$H_2(i\text{-PrGly})Cl$



$H_2(n\text{-PrGly})Cl$

Figure S2. Hydrogen bonds forming endless chains in $H_2(i\text{-PrGly})Cl$ and $H_2(n\text{-PrGly})Cl$. Hydrogen bonds are shown as dotted blue lines.

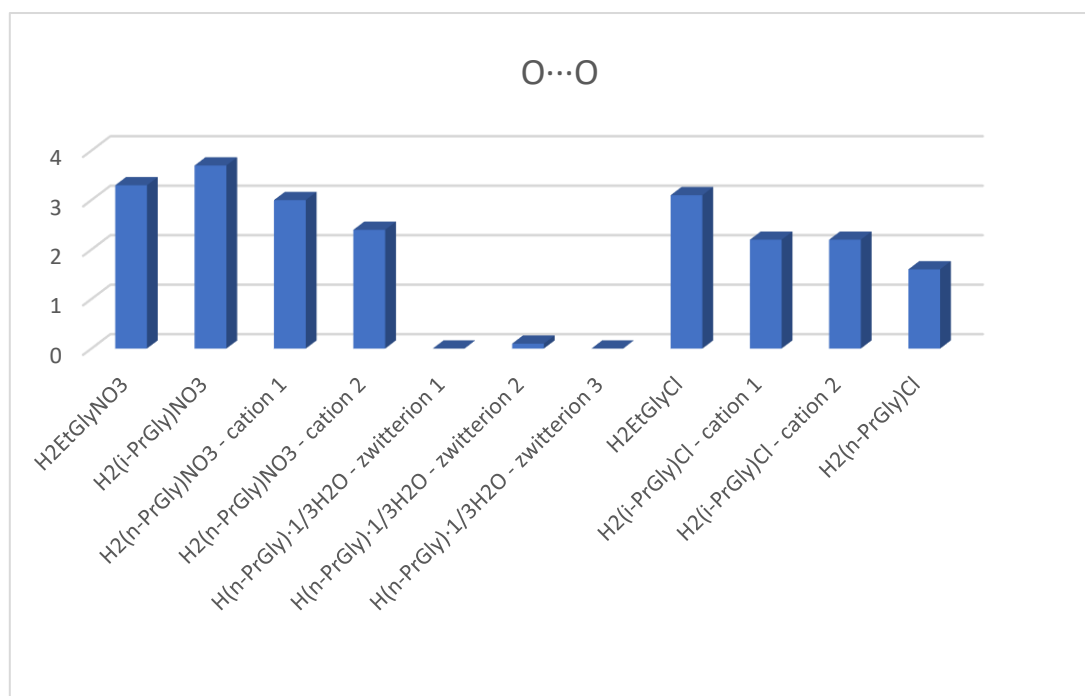


Figure S3. Fraction of O...O contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

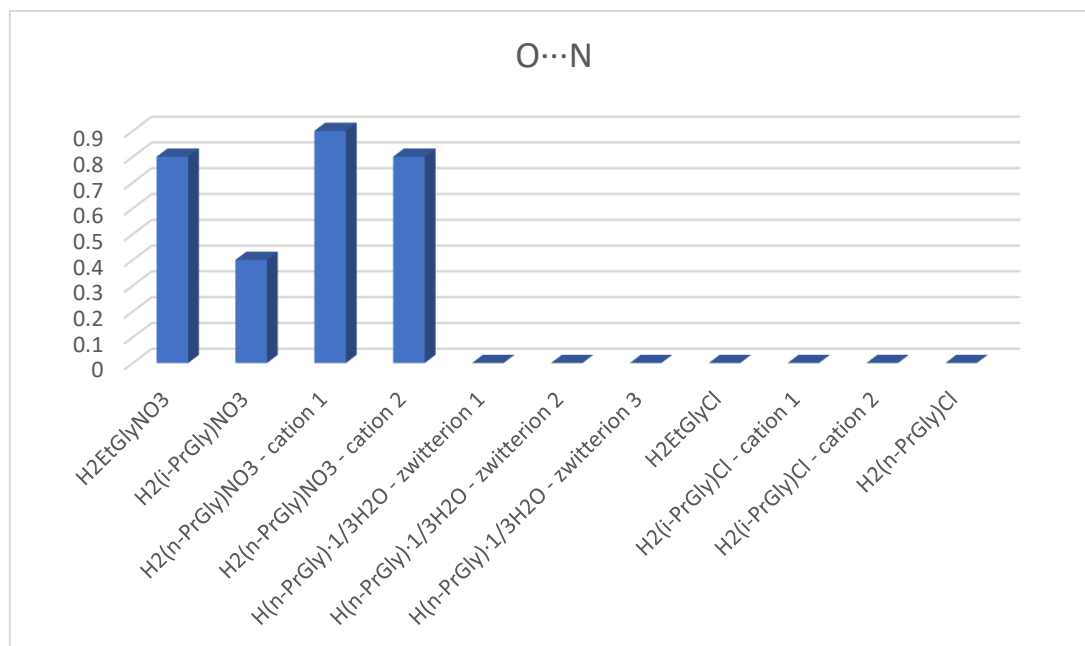


Figure S4. Fraction of O...N contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

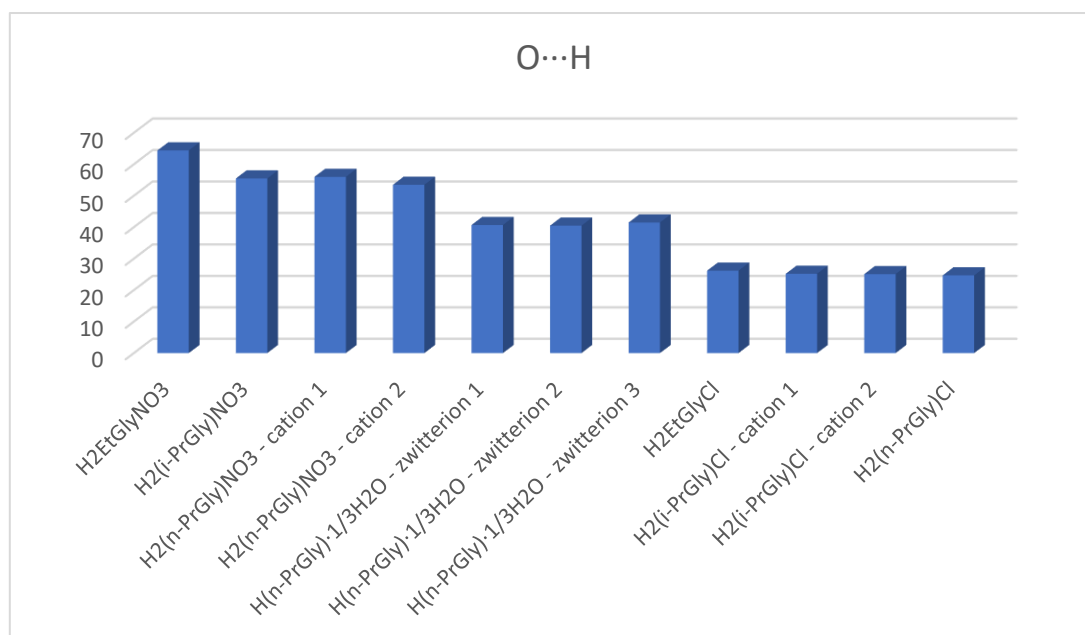


Figure S5. Fraction of O...H contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

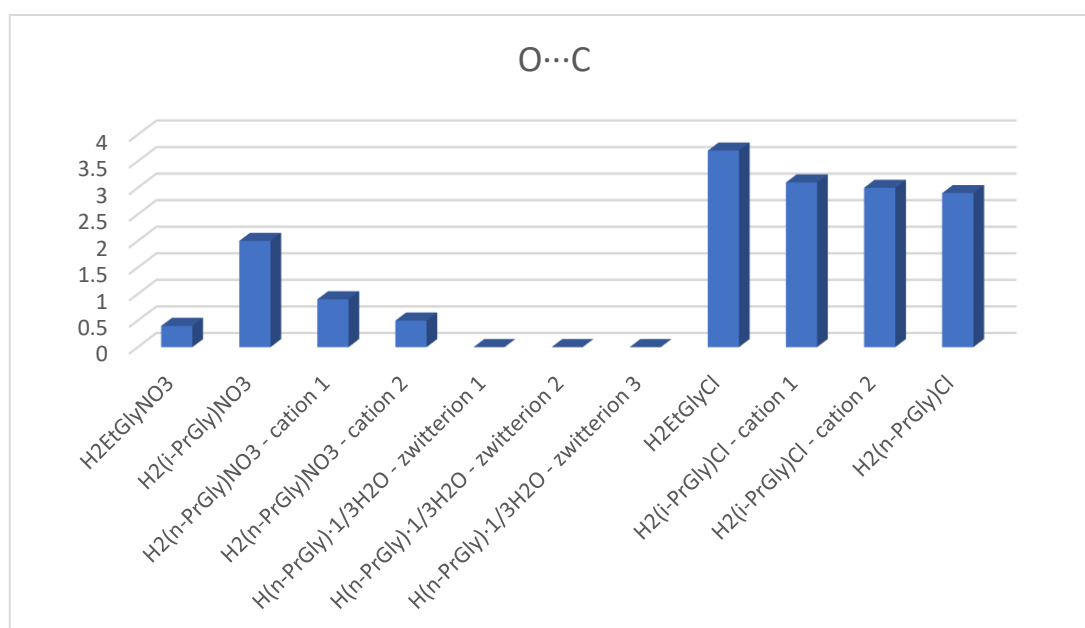


Figure S6. Fraction of O...C contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

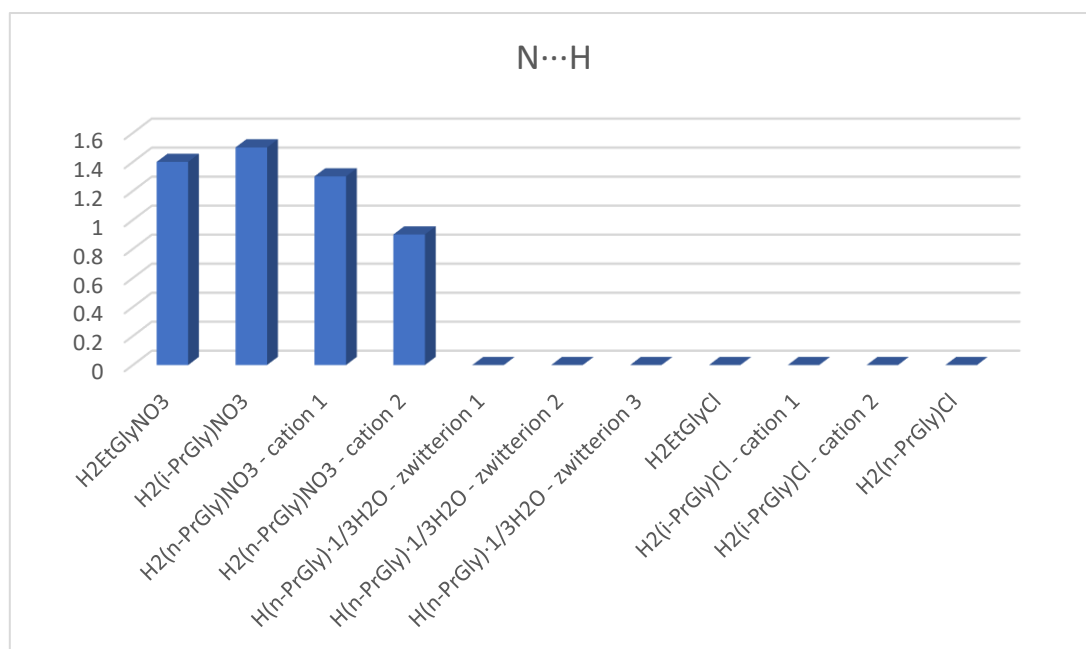


Figure S7. Fraction of N...H contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

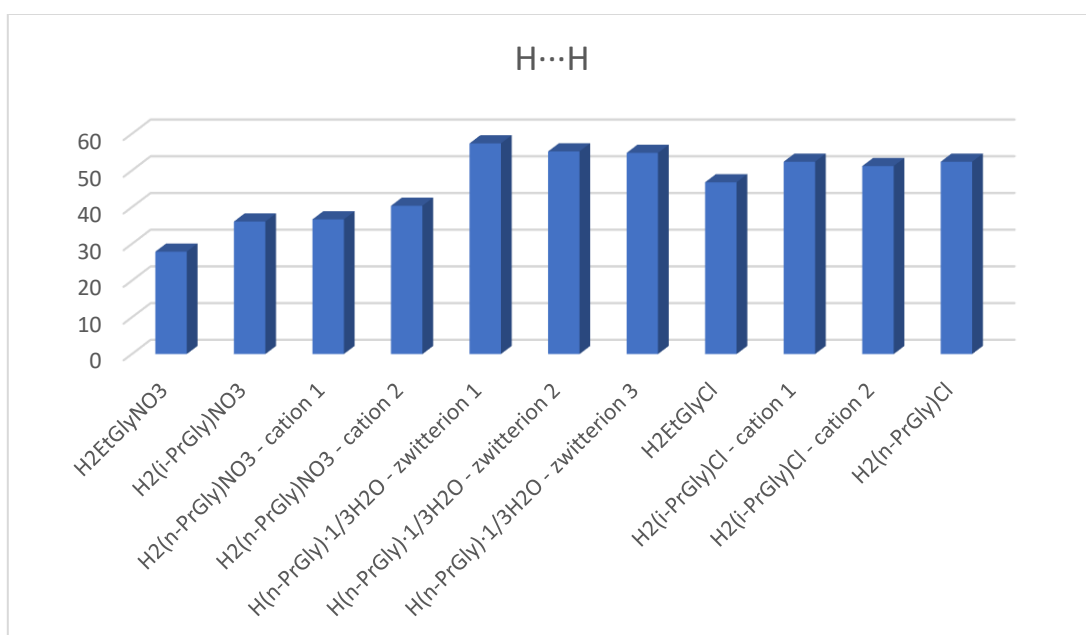


Figure S8. Fraction of H...H contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

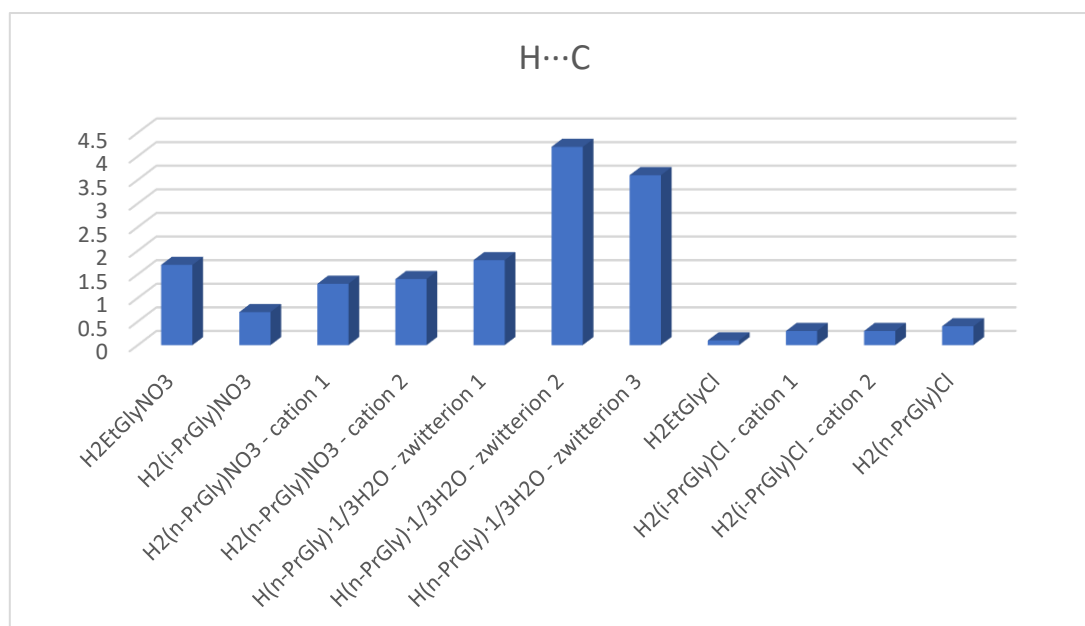


Figure S9. Fraction of H...C contacts in symmetrically independent *N*-alkylglycinium cations and zwitterions.

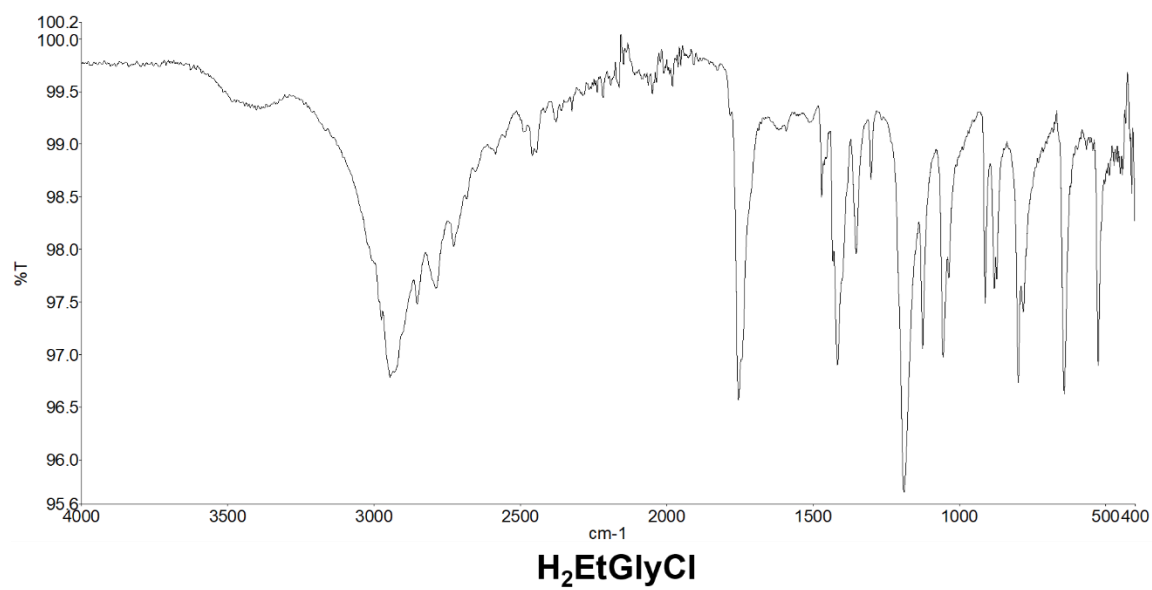


Figure S10. Infrared spectra of the $\text{H}_2\text{EtGlyCl}$.

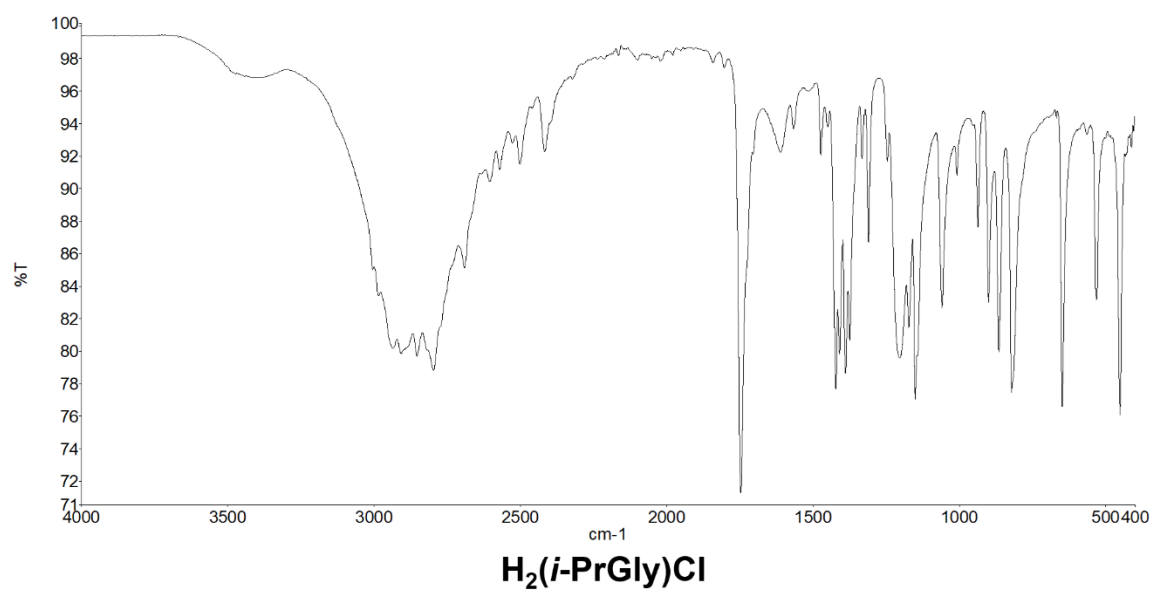


Figure S11. Infrared spectra of the $\text{H}_2(i\text{-PrGly})\text{Cl}$.

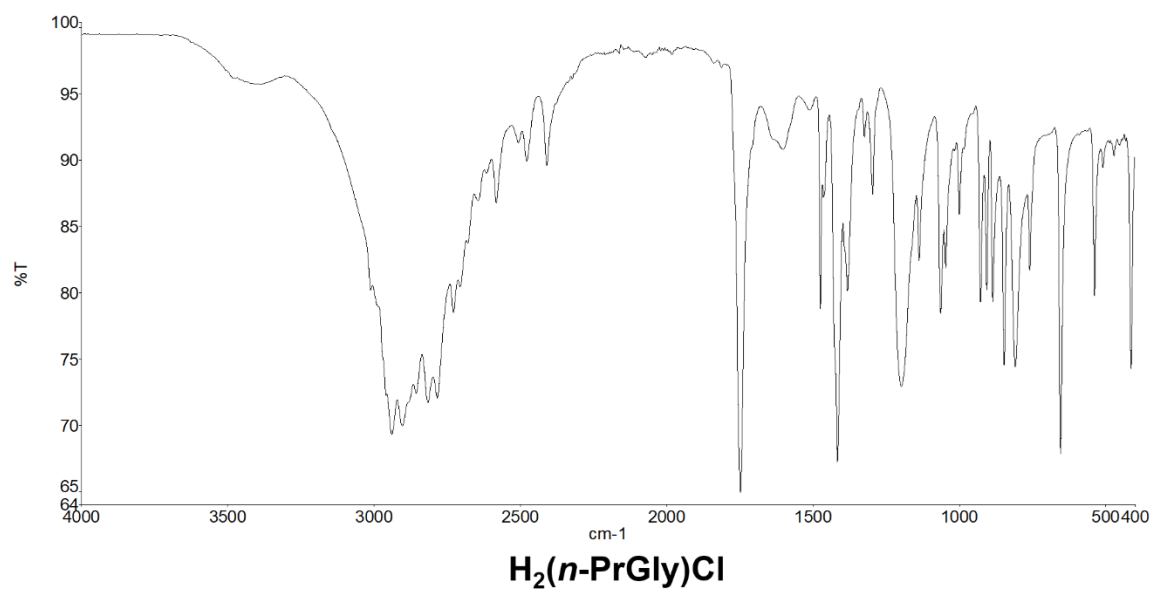


Figure S12. Infrared spectra of the H₂(*n*-PrGly)Cl.

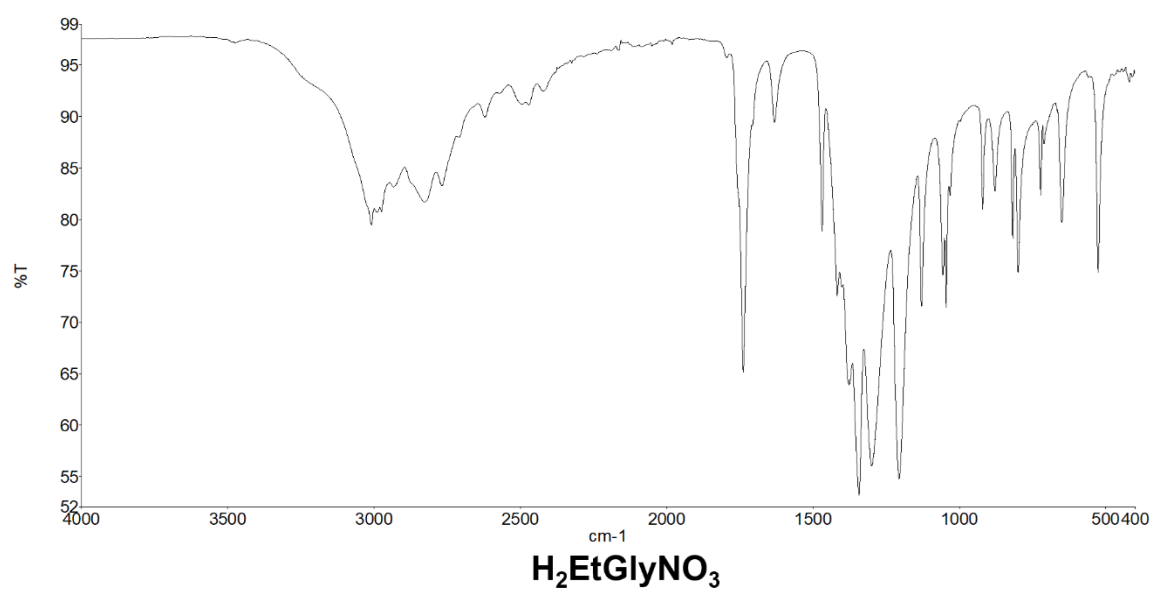


Figure S13. Infrared spectra of the H₂EtGlyNO₃.

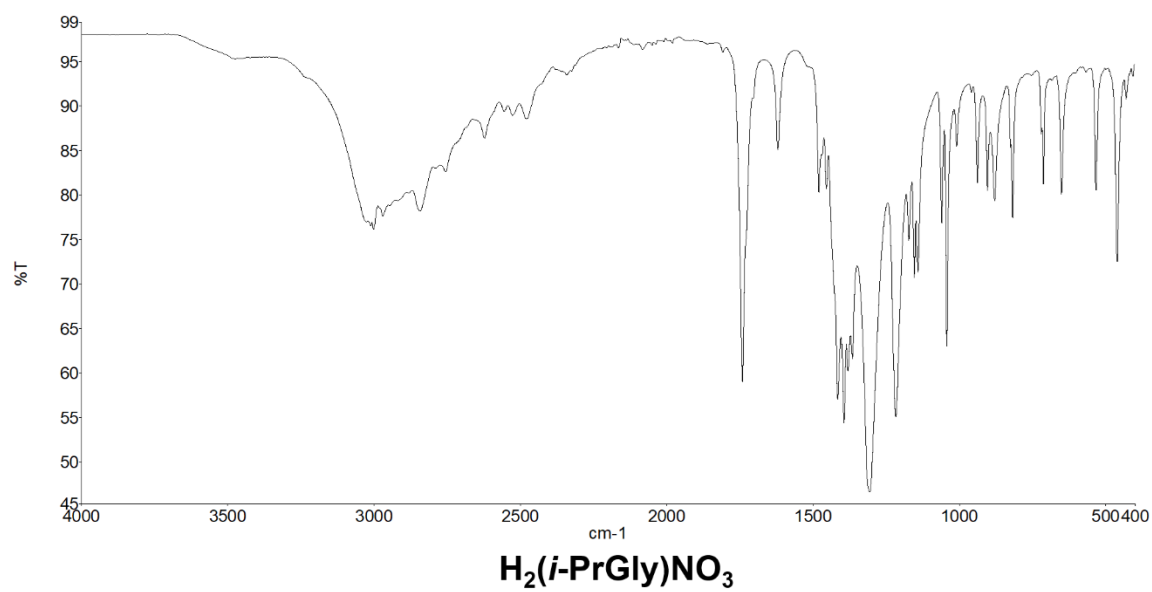


Figure S14. Infrared spectra of the $\text{H}_2(i\text{-PrGly})\text{NO}_3$.

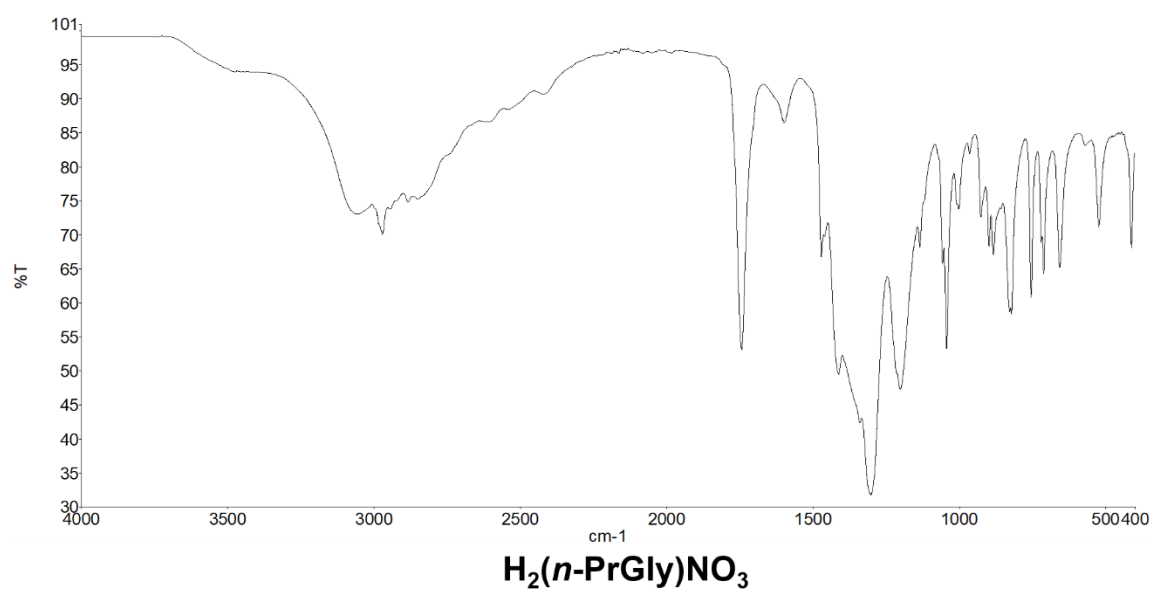


Figure S15. Infrared spectra of the $\text{H}_2(n\text{-PrGly})\text{NO}_3$.

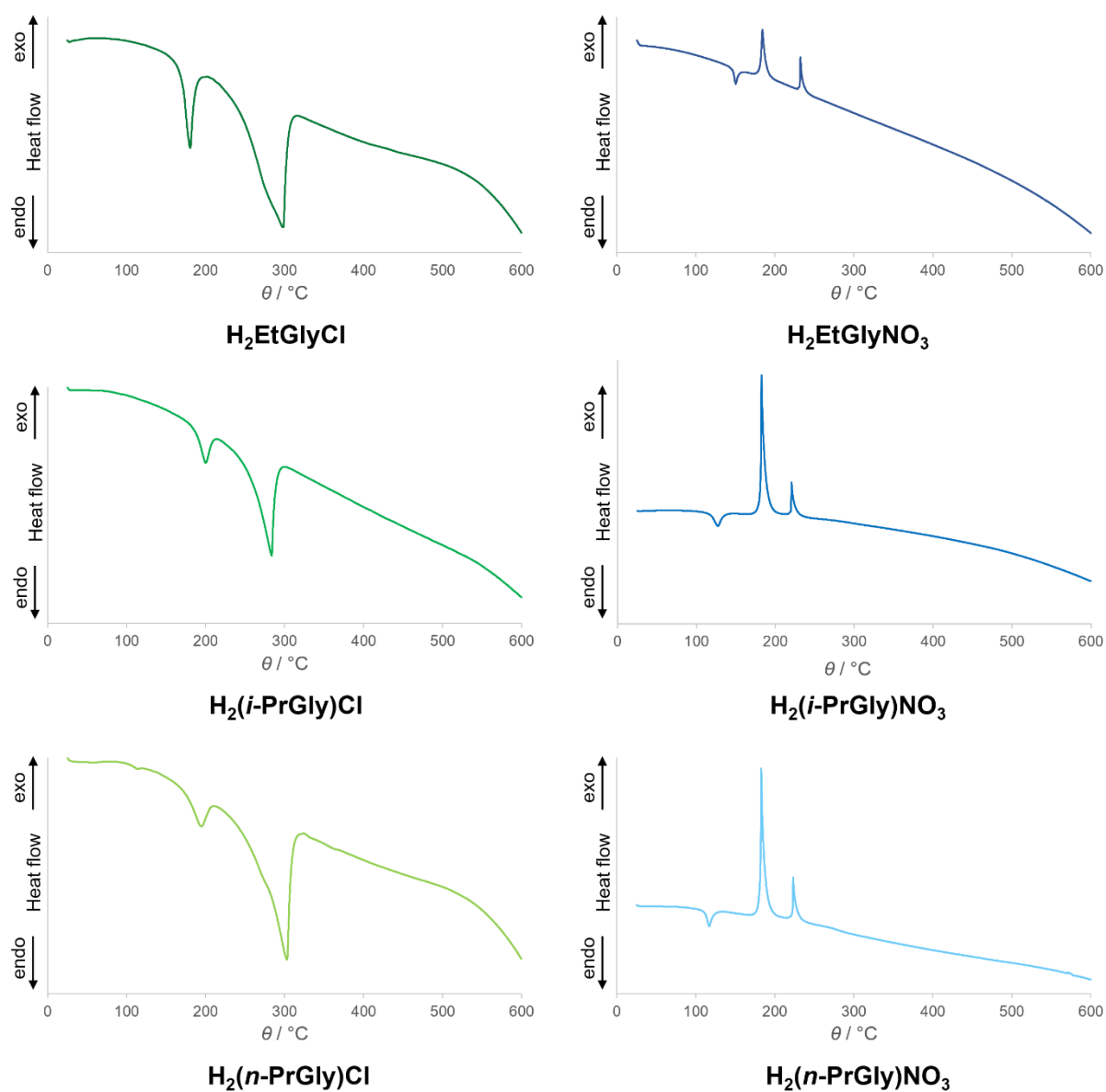


Figure S16. DSC curves of the *N*-alkylglycinium salts.

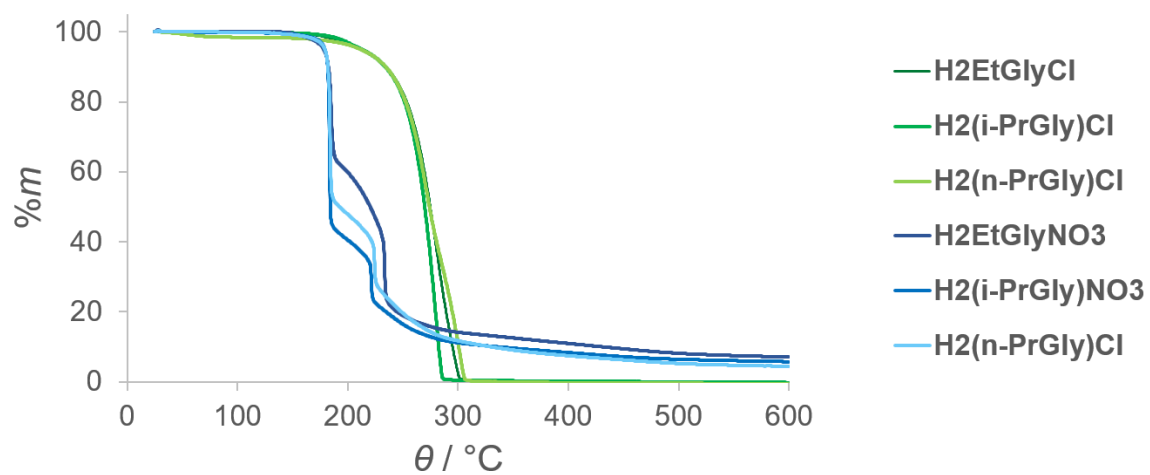


Figure S17. Thermogravimetric of the *N*-alkylglycinium salts.