

Dependence of $^1\text{H-NMR } T_1$ relaxation time of trimethylglycine betaine deep eutectic solvents on the molar composition and on the presence of water

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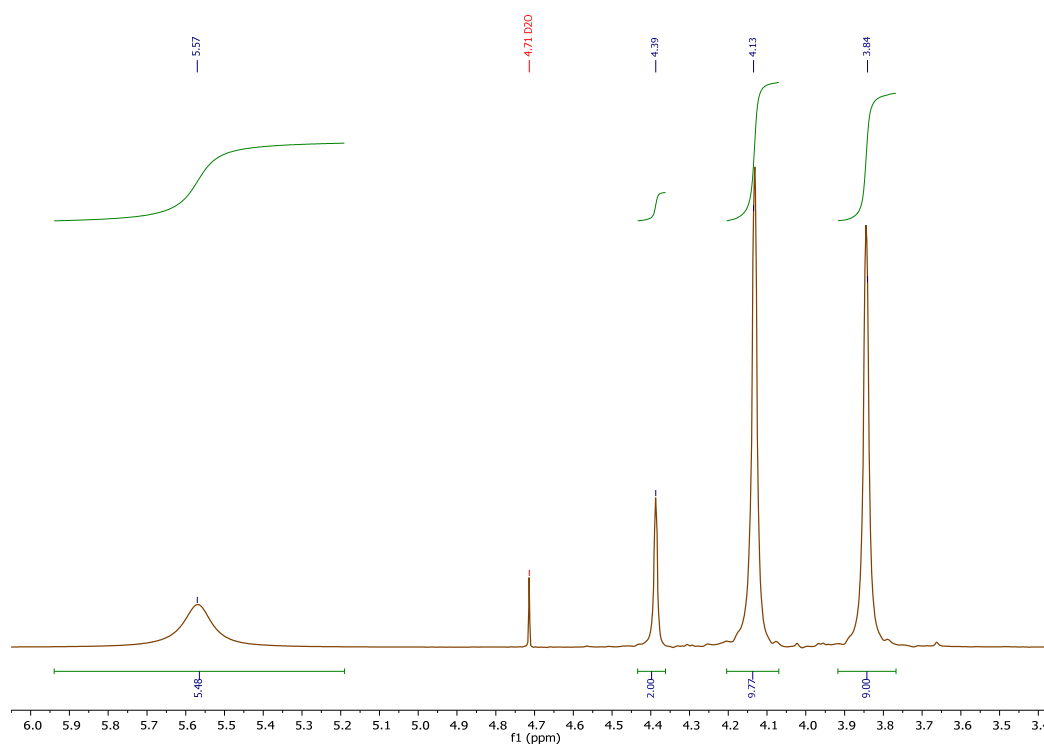
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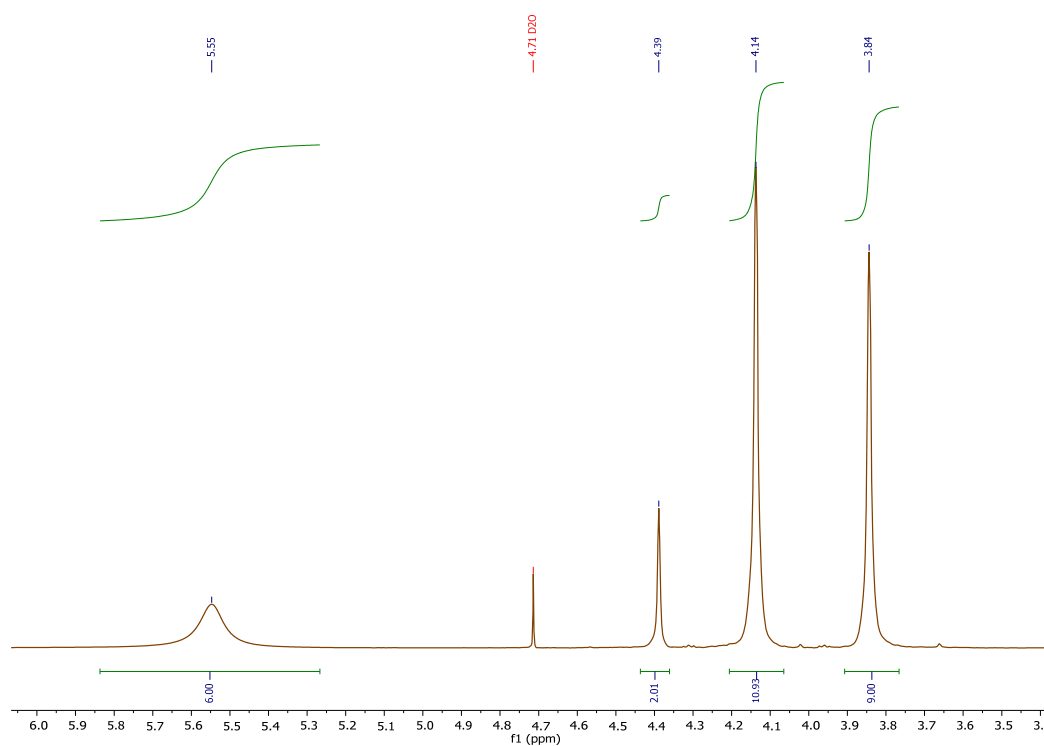
1. Determination of DES molar fraction and water content by $^1\text{H-NMR}$

1.1. $^1\text{H-NMR}$ spectra of Gb/D1 mixtures

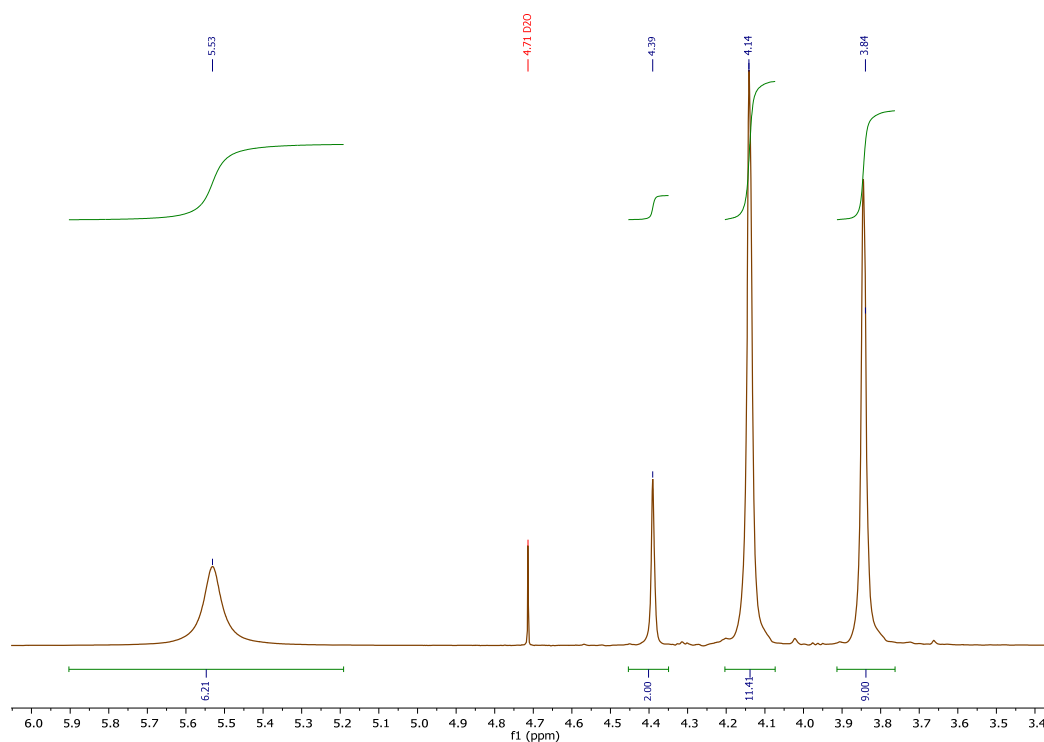
Gb/D1 1:2.44



Gb/D1 1:2.73

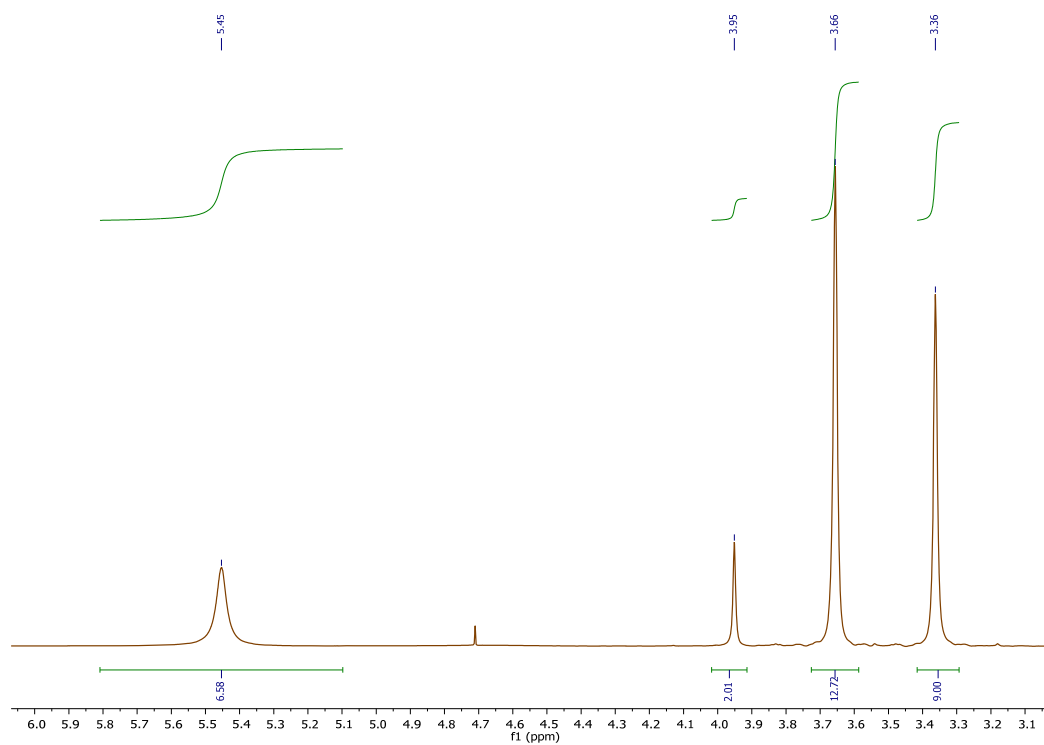


Gb/D1 1:2.85

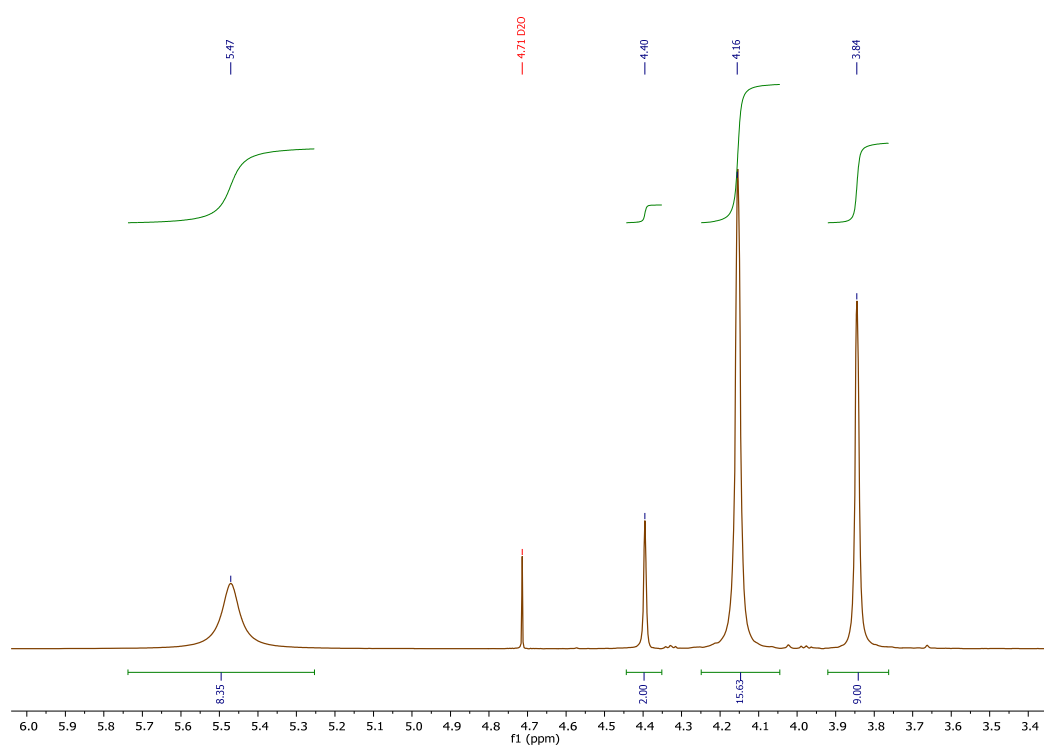


¹H-NMR (400 MHz): δ : 5.53 (s, OH, 2.85*2H), 4.39 (s, CH₂, 2H), 4.14 (s, CH₂-OH, 2.85*4H), 3.84 (s, NME₃, 9H)

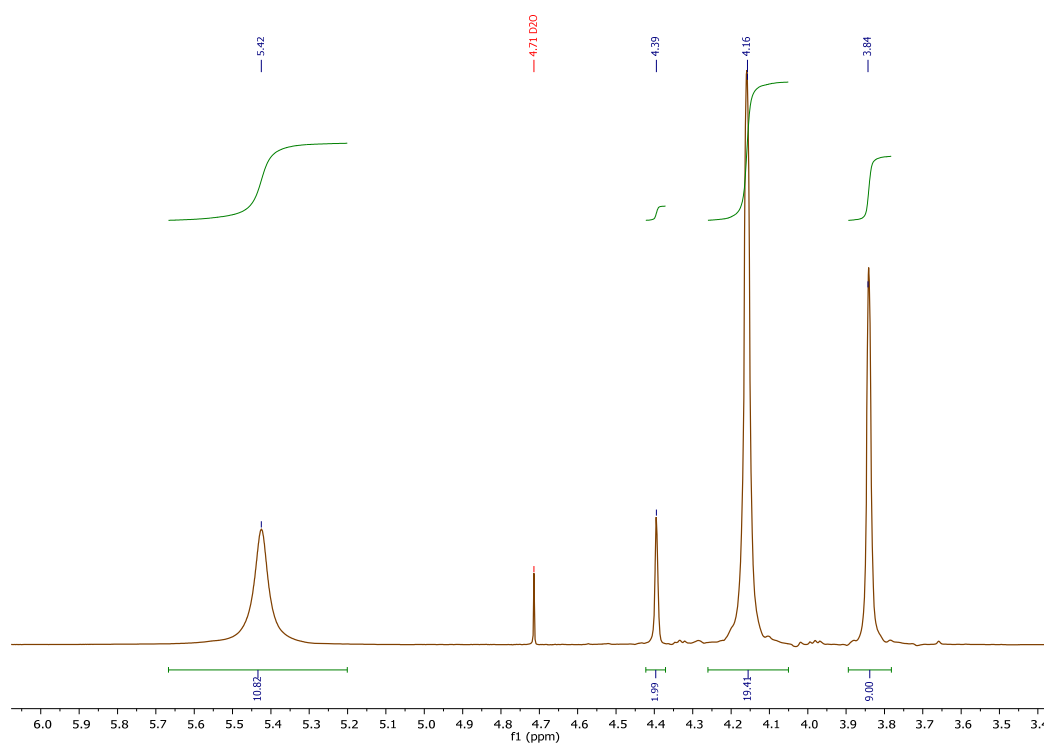
Gb/D1 1:3.18



¹H-NMR (400 MHz): δ : 5.45 (s, OH, 3.18*2H), 3.95 (s, CH₂, 2H), 3.66 (s, CH₂-OH, 3.18*4H), 3.36 (s, NME₃, 9H)

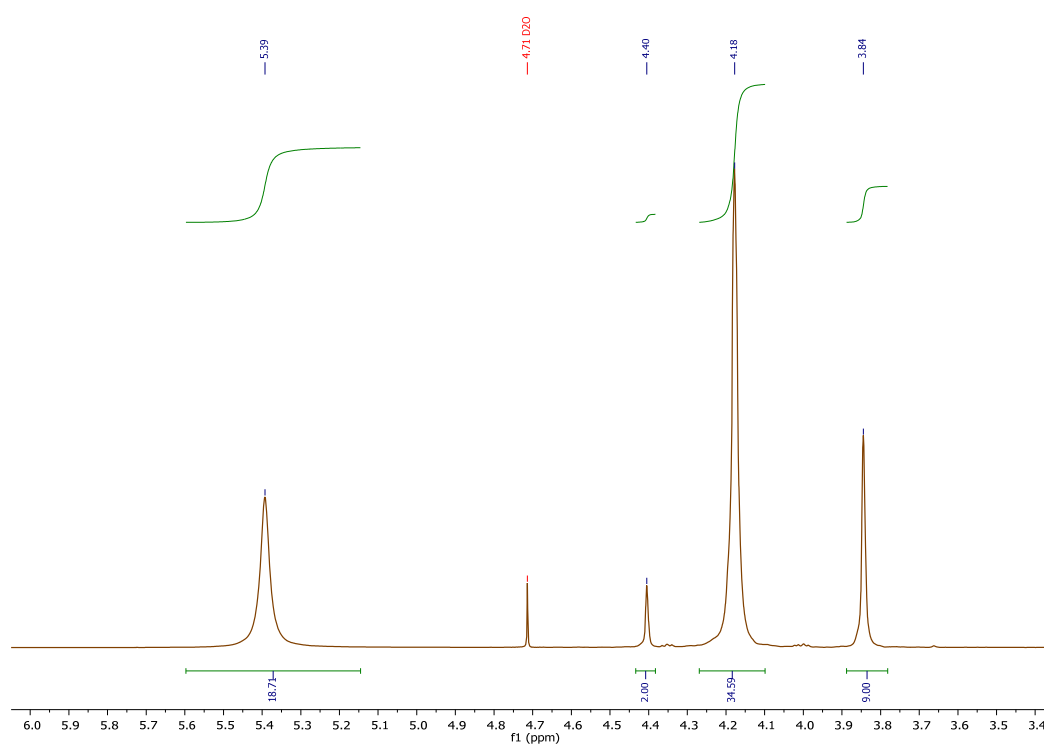
Gb/D1 1:3.91

¹H-NMR (400 MHz): δ : 5.47 (s, OH, 3.91*2H), 4.40 (s, CH₂, 2H), 4.16 (s, CH₂-OH, 3.91*4H), 3.84 (s, NME₃, 9H)

Gb/D1 1:4.85

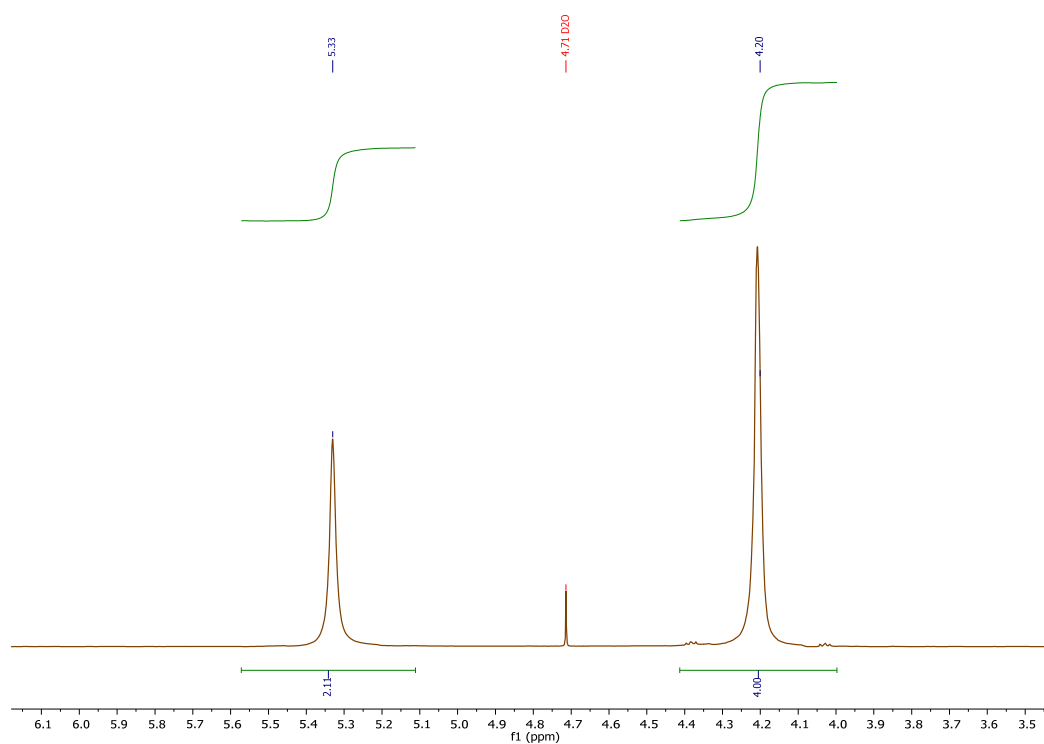
¹H-NMR (400 MHz): δ : 5.42 (s, OH, 4.85*2H), 4.39 (s, CH₂, 2H), 4.16 (s, CH₂-OH, 4.85*4H), 3.84 (s, NME₃, 9H)

Gb/D1 1:8.65

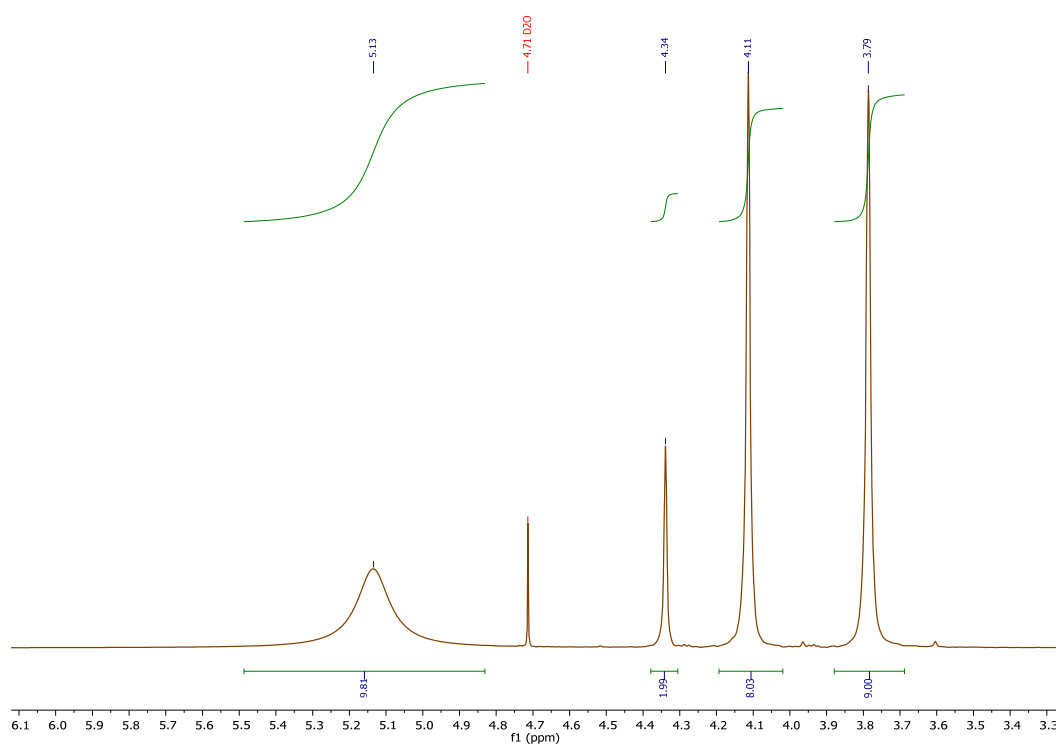


¹H-NMR (400 MHz): δ : 5.39 (s, OH, 8.65*2H), 4.40 (s, CH₂, 2H), 4.18 (s, CH₂-OH, 8.65*4H), 3.84 (s, NME₃, 9H)

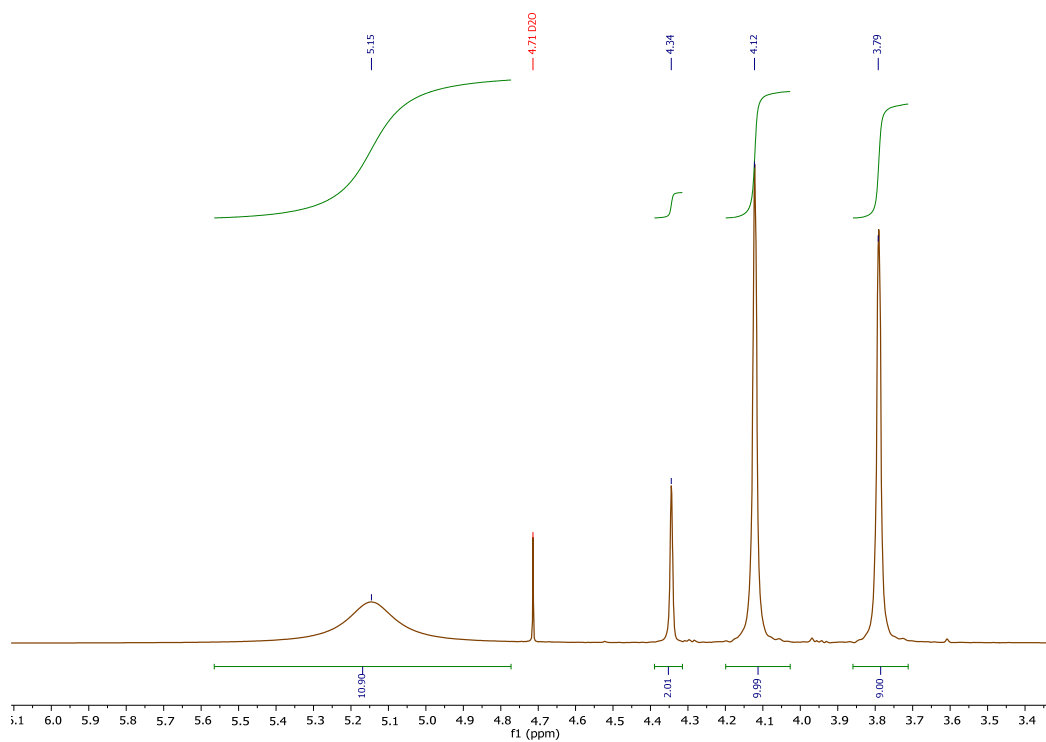
D1



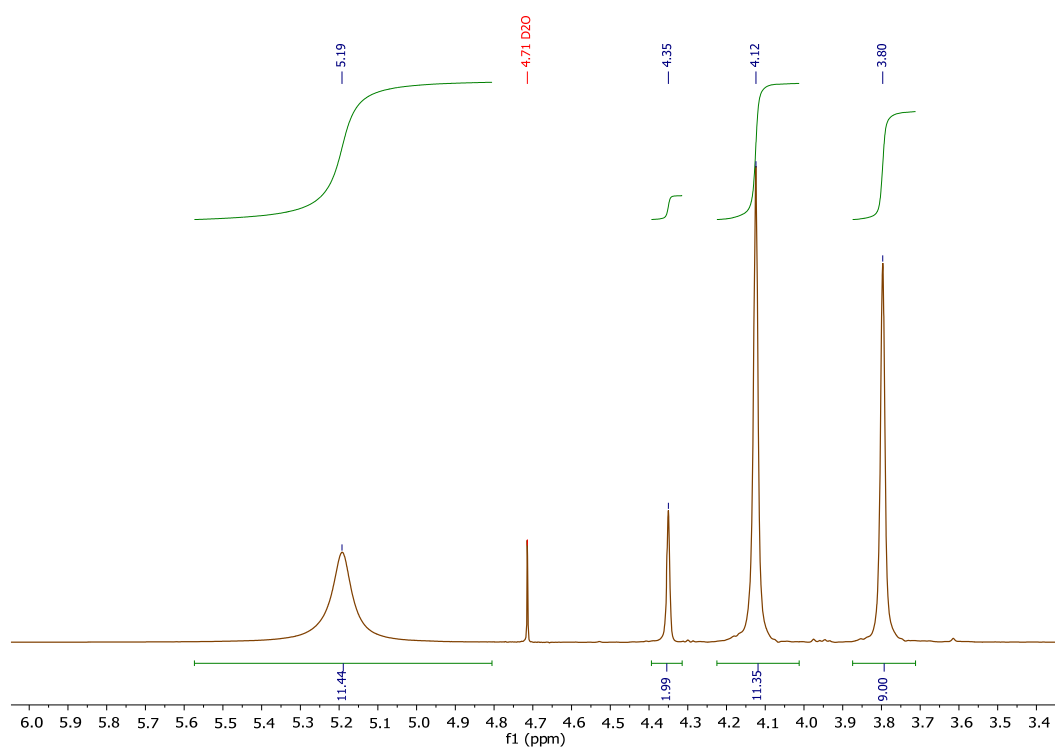
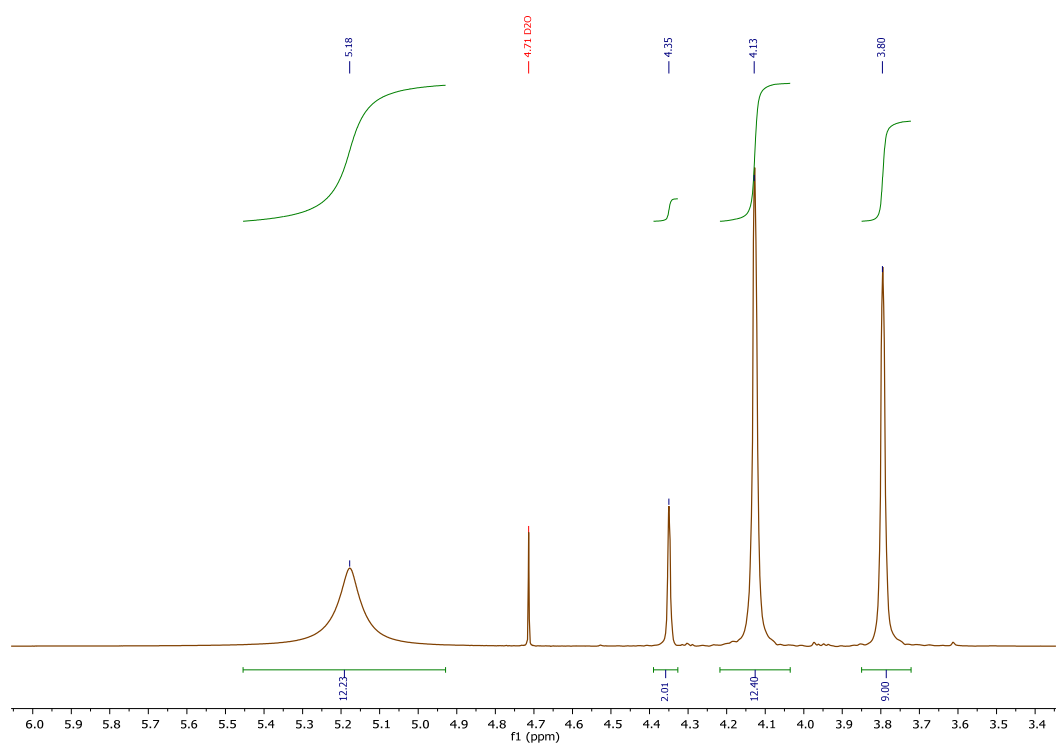
¹H-NMR (400 MHz): δ : 5.33 (s, OH), 4.20 (s, CH₂-OH, 4H)

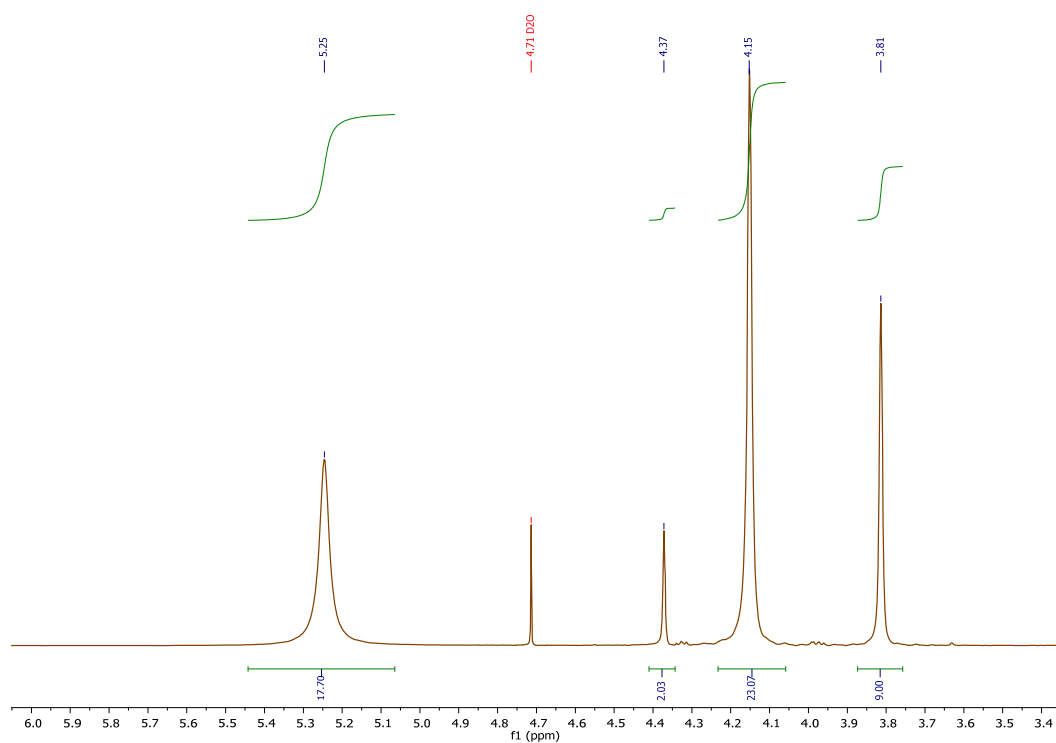
1.2. $^1\text{H-NMR}$ spectra of Gb/D1 mixtures + 3 eq H_2O Gb/D1 1:2.01 + 3 eq H_2O 

$^1\text{H-NMR}$ (400 MHz): δ : 5.13 (s, OH, 2.01*2H), 4.34 (s, CH_2 , 2H), 4.11 (s, $\text{CH}_2\text{-OH}$, 2.01*4H), 3.79 (s, NME_3 , 9H)

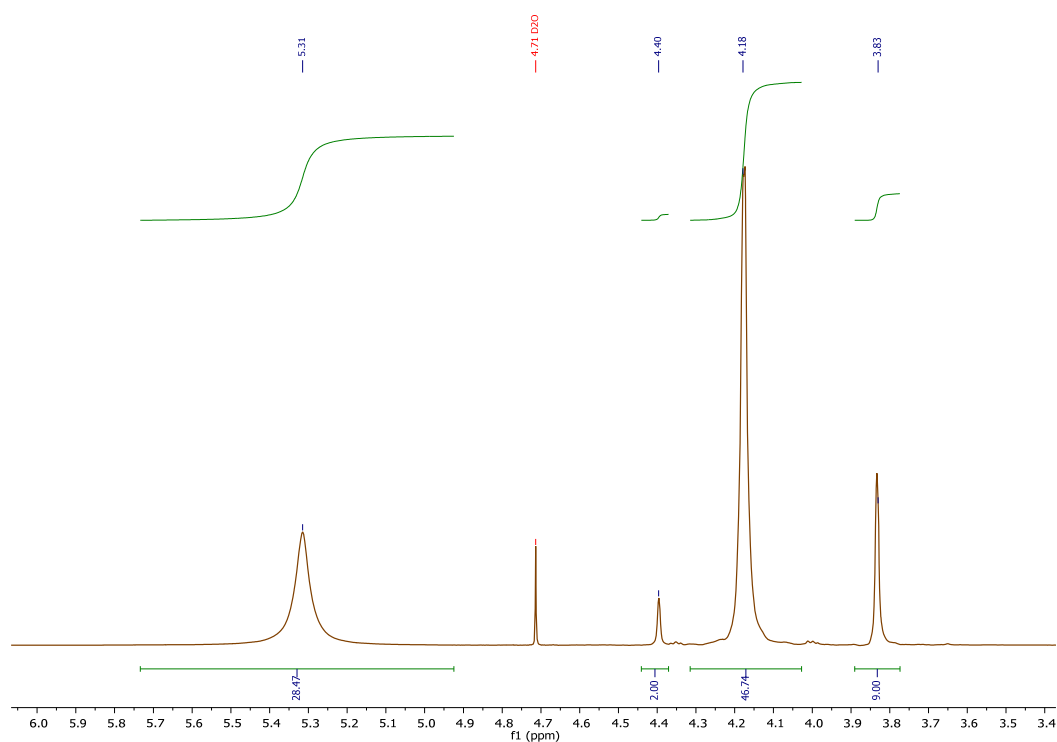
Gb/D1 1:2.49 + 3 eq H_2O 

$^1\text{H-NMR}$ (400 MHz): δ : 5.15 (s, OH, 2.49*2H), 4.34 (s, CH_2 , 2H), 4.12 (s, $\text{CH}_2\text{-OH}$, 2.49*4H), 3.79 (s, NME_3 , 9H)

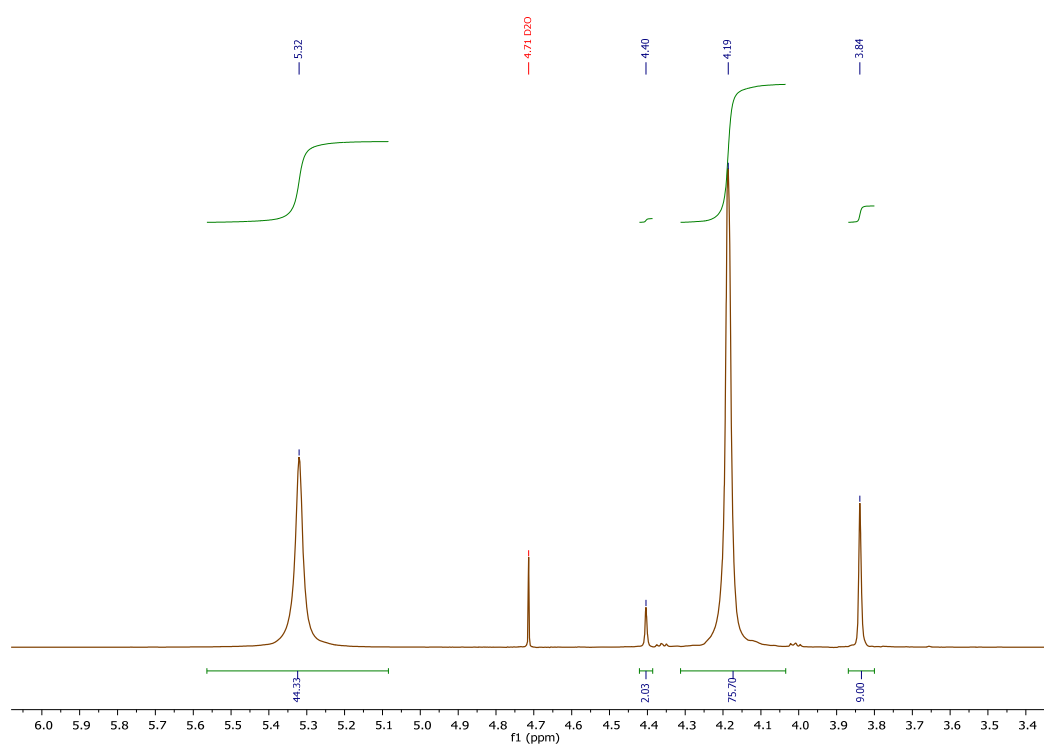
Gb/D1 1:2.85 + 3 eq H₂O**Gb/D1 1:3.11 + 3 eq H₂O**

Gb/D1 1:5.76 + 3 eq H₂O

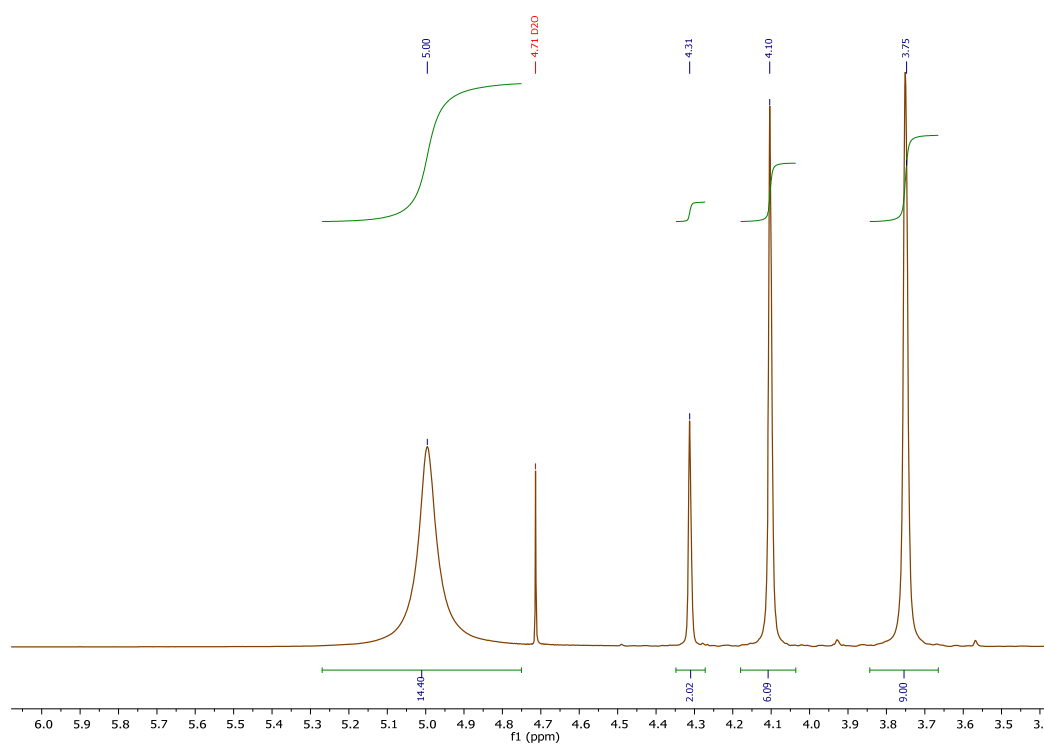
¹H-NMR (400 MHz): δ : 5.25 (s, OH, 5.76*2H), 4.37 (s, CH₂, 2H), 4.15 (s, CH₂-OH, 5.76*4H), 3.81 (s, NME₃, 9H)

Gb/D1 1:11.81 + 3 eq H₂O

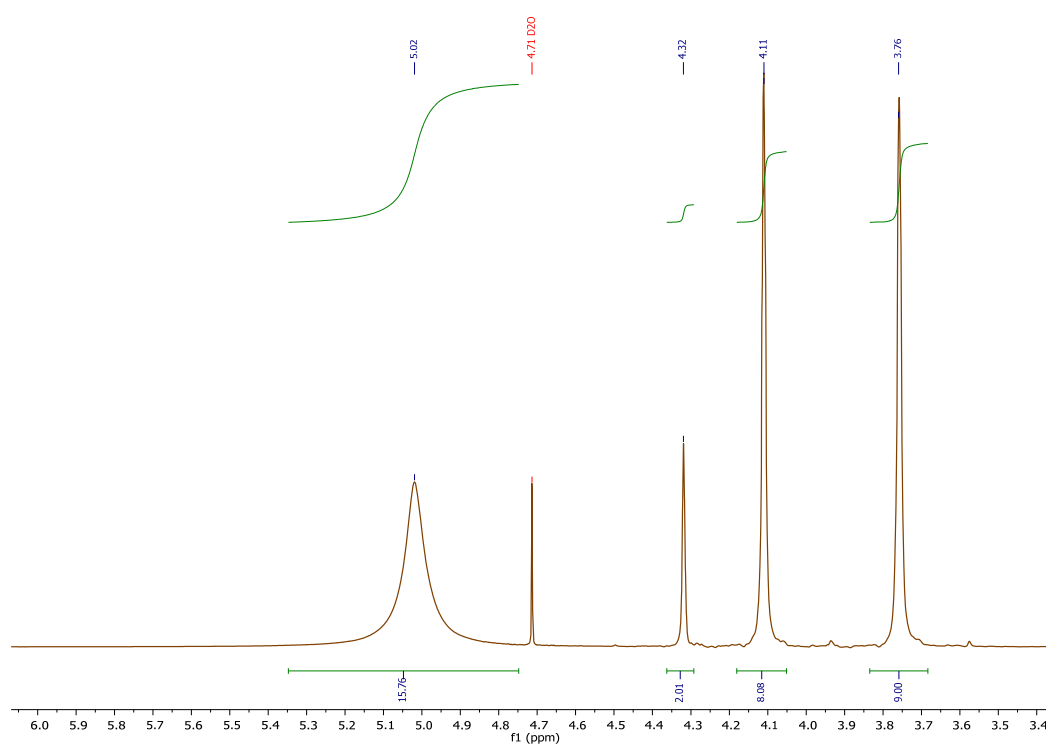
¹H-NMR (400 MHz): δ : 5.31 (s, OH, 11.81*2H), 4.40 (s, CH₂, 2H), 4.18 (s, CH₂-OH, 11.81*4H), 3.83 (s, NME₃, 9H)

Gb/D1 1:18.99 + 3 eq H₂O

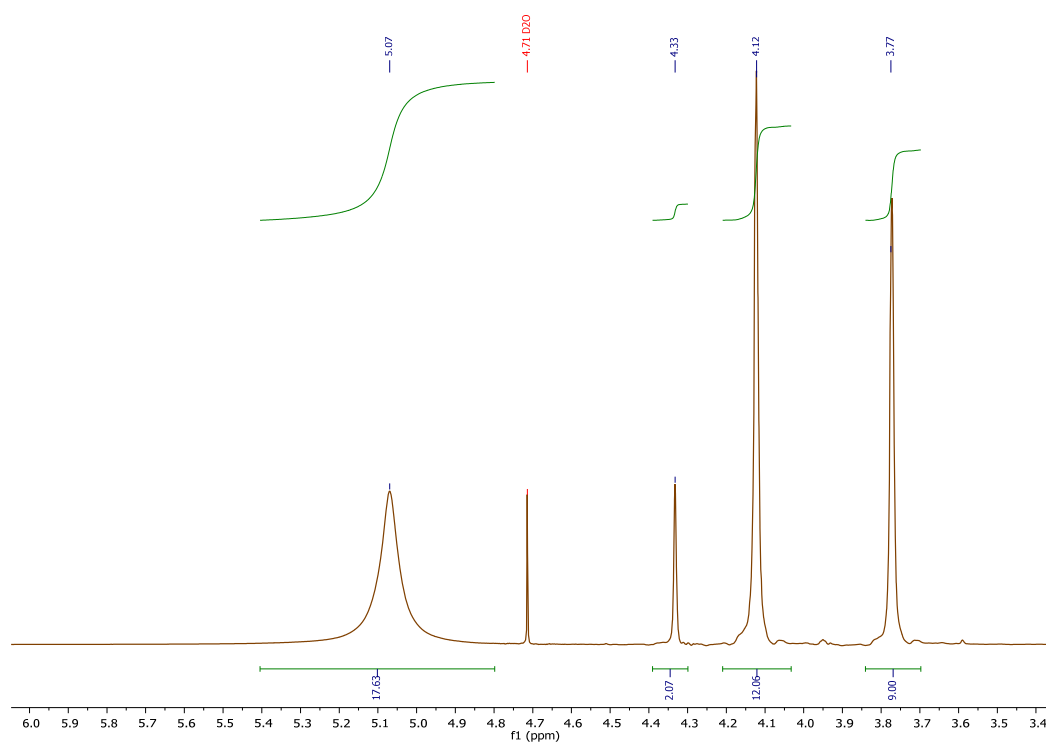
¹H-NMR (400 MHz): δ : 5.32 (s, OH, 18.99*2H), 4.40 (s, CH₂, 2H), 4.19 (s, CH₂-OH, 18.99*4H), 3.84 (s, NME₃, 9H)

1.3. ¹H-NMR spectra of Gb/D1 mixtures + 6 eq H₂O**Gb/D1 1:1.52 + 6 eq H₂O**

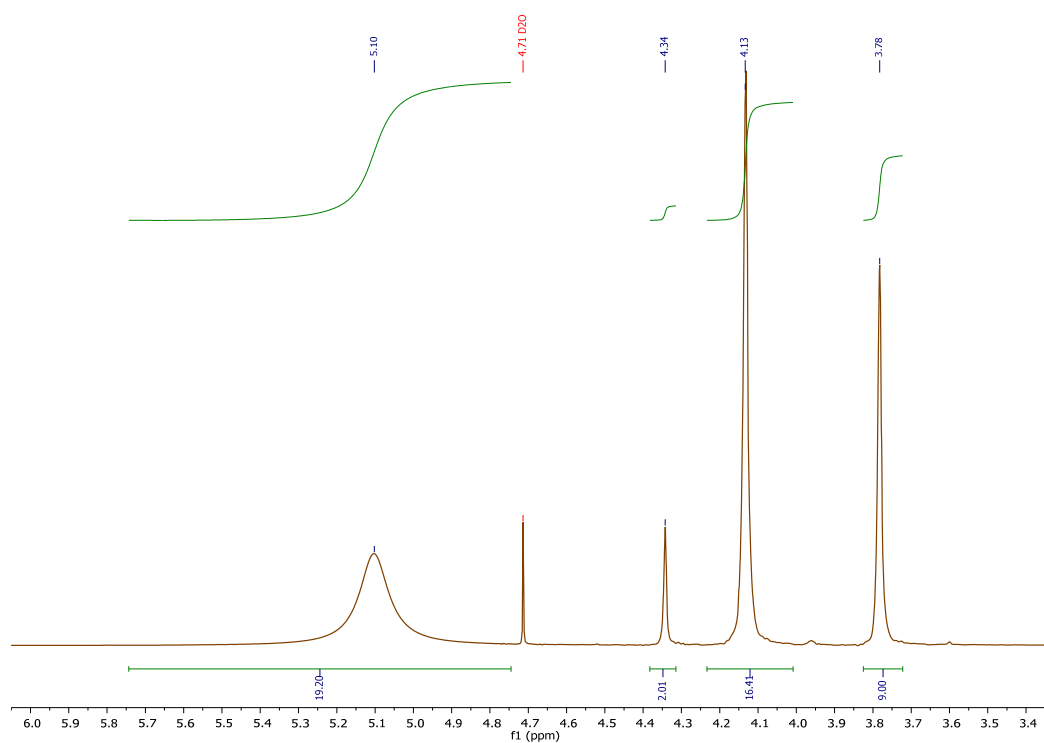
¹H-NMR (400 MHz): δ : 5.00 (s, OH, 1.52*2H), 4.31 (s, CH₂, 2H), 4.10 (s, CH₂-OH, 1.52*4H), 3.75 (s, NME₃, 9H)

Gb/D1 1:2.02 + 6 eq H₂O

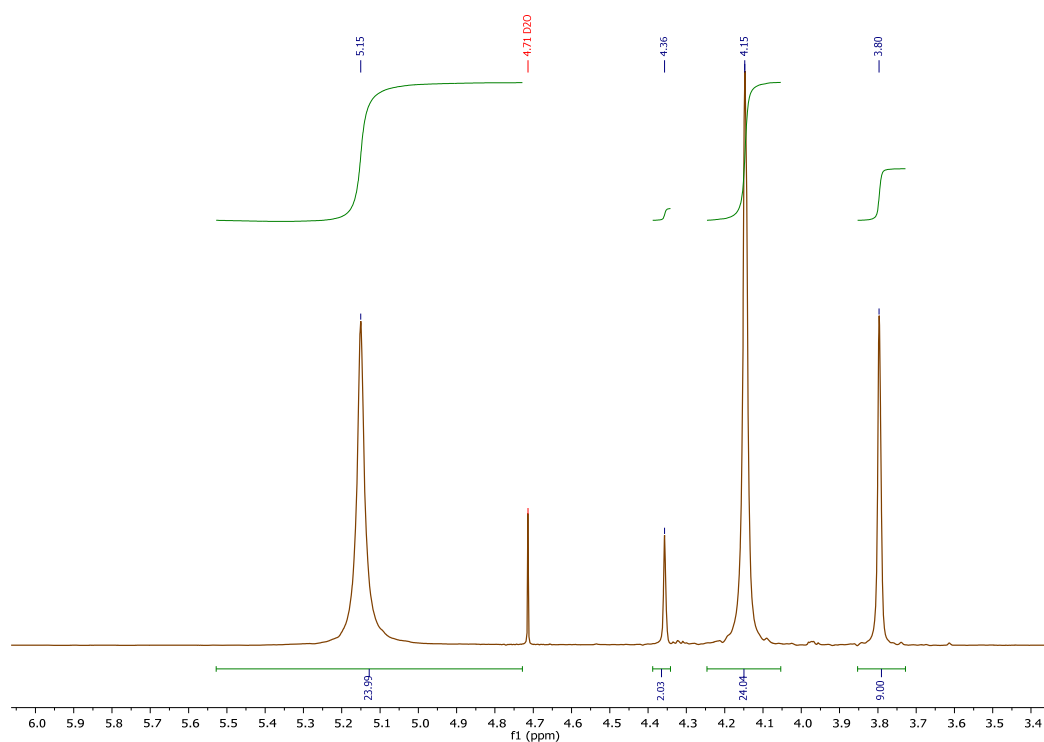
¹H-NMR (400 MHz): δ : 5.02 (s, OH, 2.02*2H), 4.32 (s, CH₂, 2H), 4.11 (s, CH₂-OH, 2.02*4H), 3.76 (s, NME₃, 9H)

Gb/D1 1:3.02 + 6 eq H₂O

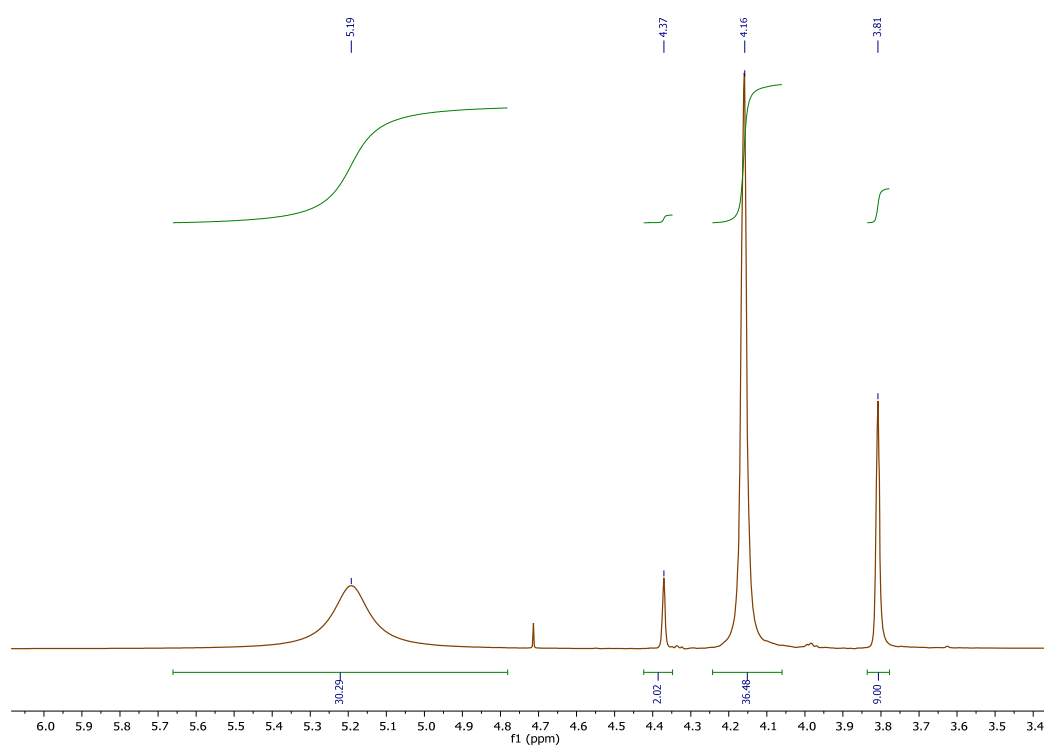
¹H-NMR (400 MHz): δ : 5.07 (s, OH, 3.02*2H), 4.33 (s, CH₂, 2H), 4.12 (s, CH₂-OH, 3.02*4H), 3.77 (s, NME₃, 9H)

Gb/D1 1:4.10 + 6 eq H₂O

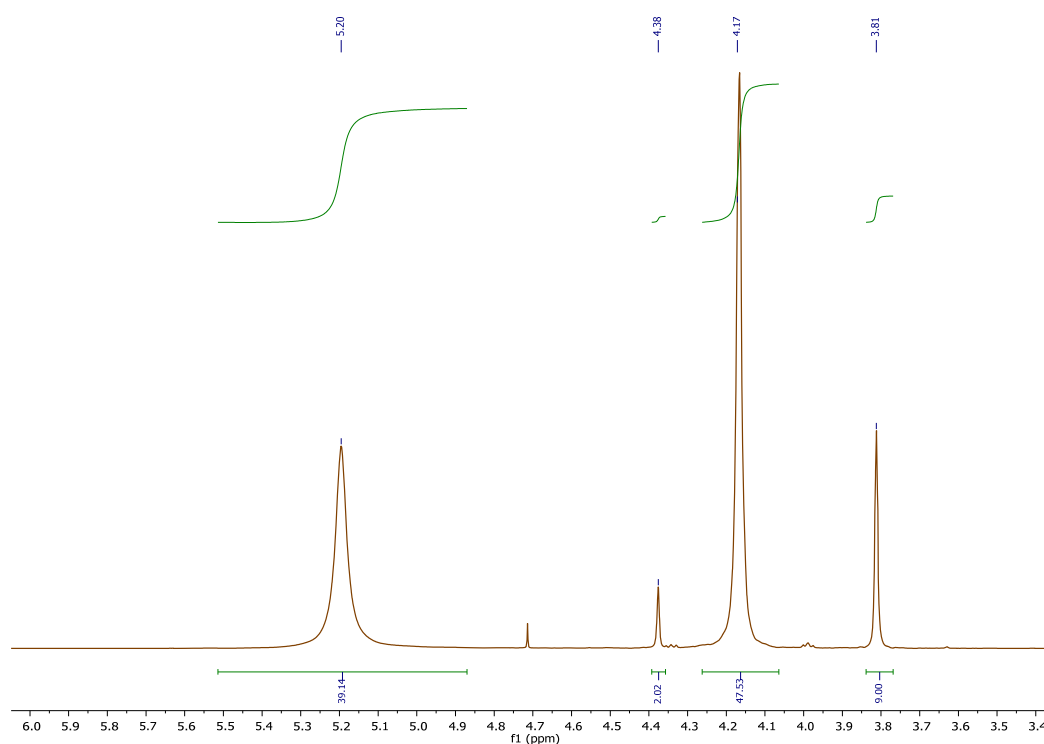
¹H-NMR (400 MHz): δ : 5.10 (s, OH, 4.10*2H), 4.34 (s, CH₂, 2H), 4.13 (s, CH₂-OH, 4.10*4H), 3.78 (s, NME₃, 9H)

Gb/D1 1:6.05 + 6 eq H₂O

¹H-NMR (400 MHz): δ : 5.15 (s, OH, 6.05*2H), 4.36 (s, CH₂, 2H), 4.15 (s, CH₂-OH, 6.05*4H), 3.80 (s, NME₃, 9H)

Gb/D1 1:9.11 + 6 eq H₂O

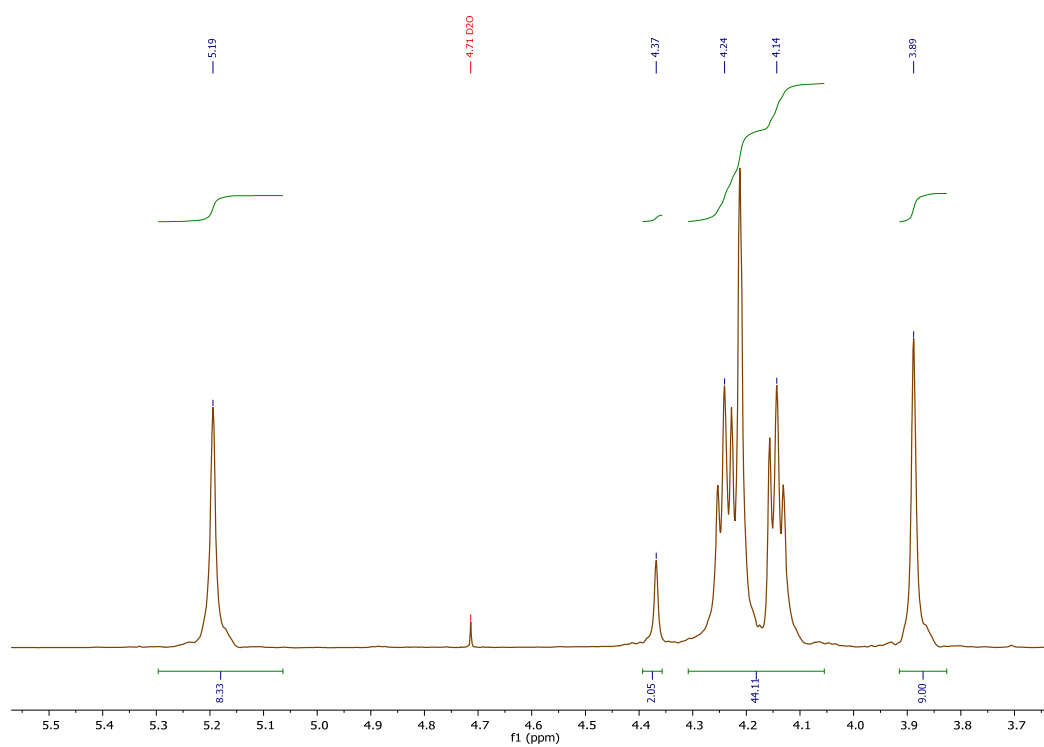
¹H-NMR (400 MHz): δ : 5.19 (s, OH, 9.11*2H), 4.37 (s, CH₂, 2H), 4.16 (s, CH₂-OH, 9.11*4H), 3.81 (s, NME₃, 9H)

Gb/D1 1:11.82 + 6 eq H₂O

¹H-NMR (400 MHz): δ : 5.20 (s, OH, 11.82*2H), 4.38 (s, CH₂, 2H), 4.17 (s, CH₂-OH, 11.82*4H), 3.81 (s, NME₃, 9H)

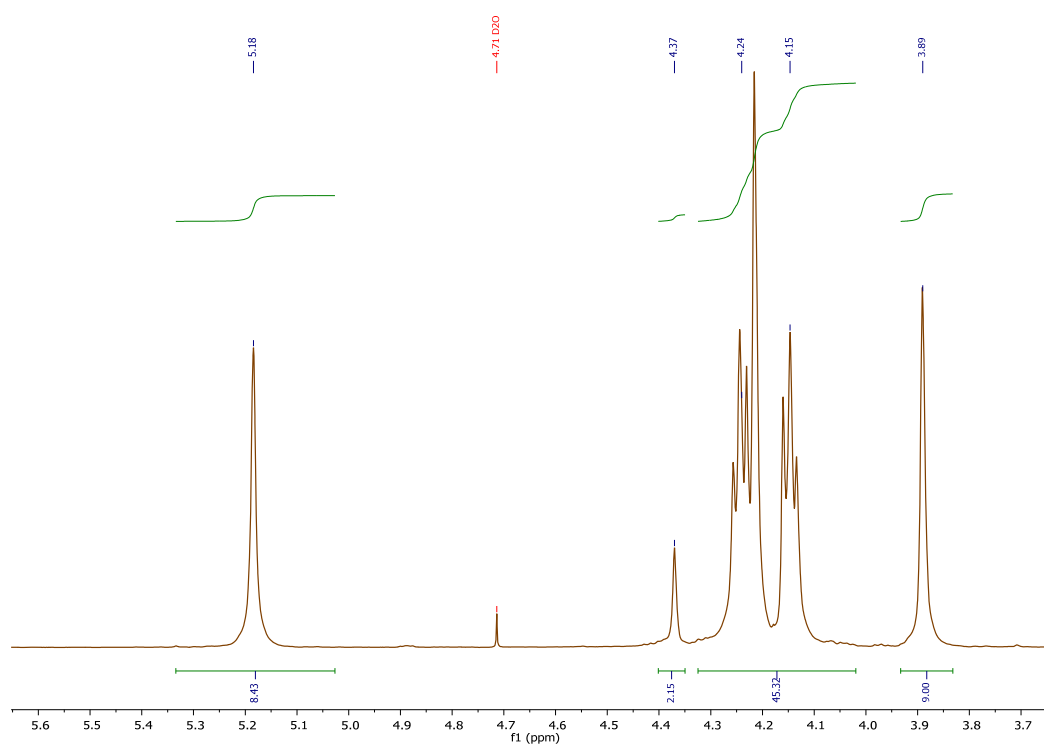
1.4. $^1\text{H-NMR}$ spectra of Gb/D2 mixtures

Gb/D2 1:3.66



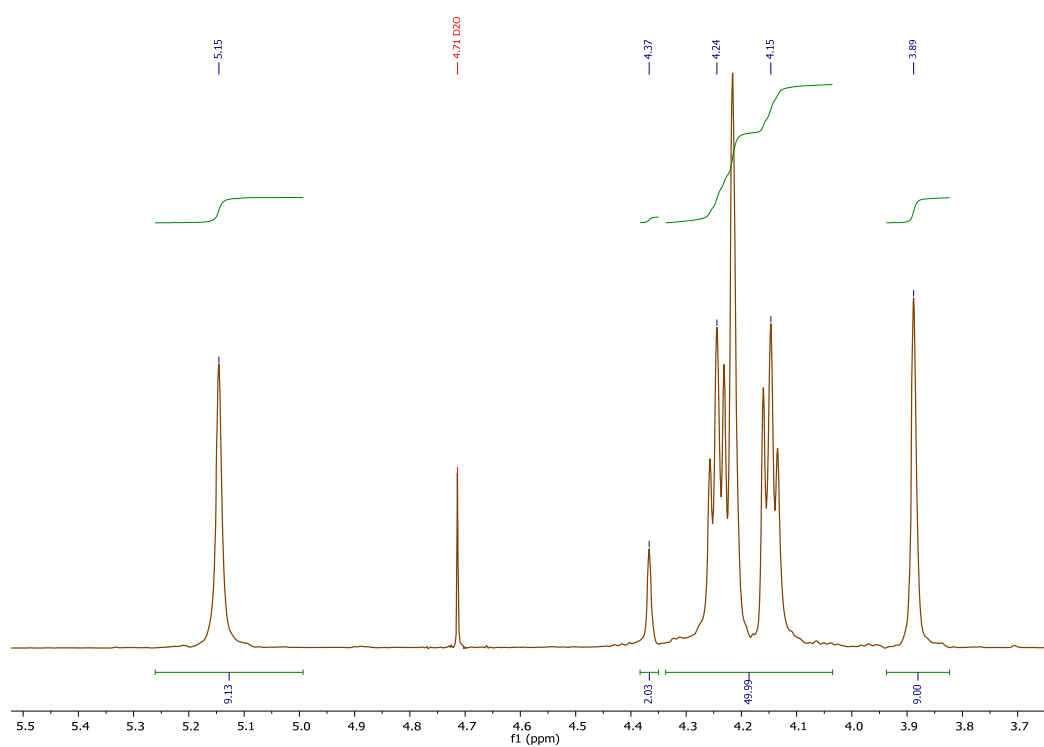
$^1\text{H-NMR}$ (400 MHz): δ : 5.19 (s, OH, 3.66*2H), 4.37 (s, CH_2 , 2H), 4.24-4.14 (m, $\text{CH}_2\text{-OH}$ and $\text{CH}_2\text{-O-CH}_2$, 3.66*12H), 3.84 (s, NME_3 , 9H)

Gb/D2 1:3.76



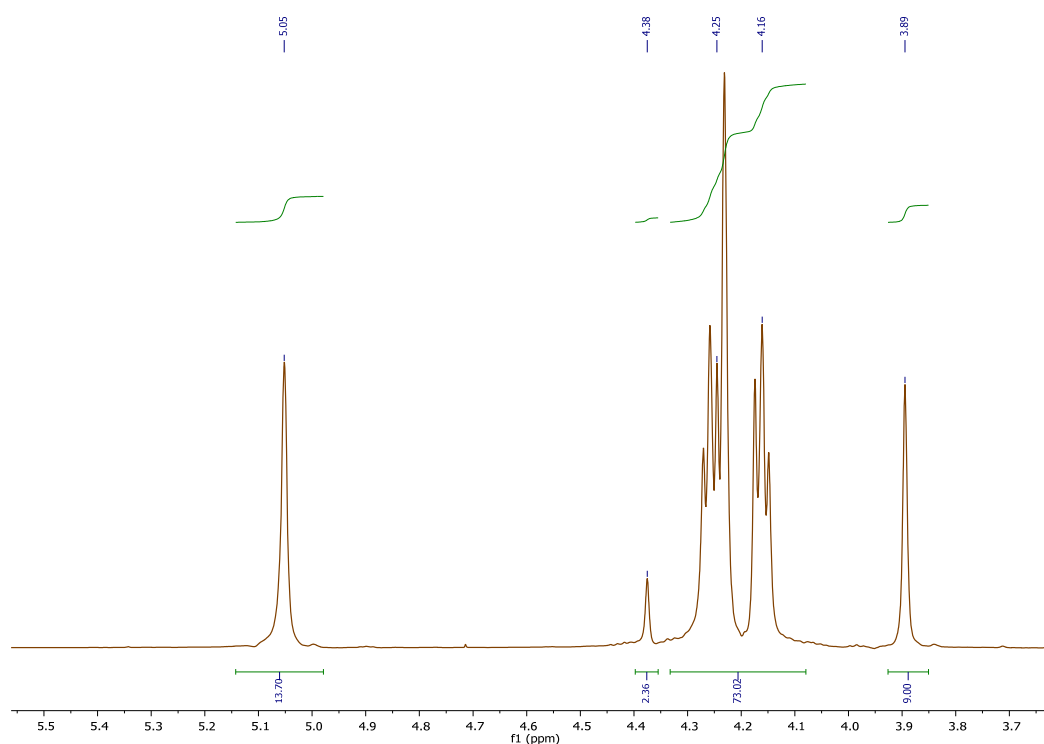
$^1\text{H-NMR}$ (400 MHz): δ : 5.18 (s, OH, 3.76*2H), 4.37 (s, CH_2 , 2H), 4.24-4.15 (m, $\text{CH}_2\text{-OH}$ and $\text{CH}_2\text{-O-CH}_2$, 3.66*12H), 3.89 (s, NME_3 , 9H)

Gb/D2 1:4.21



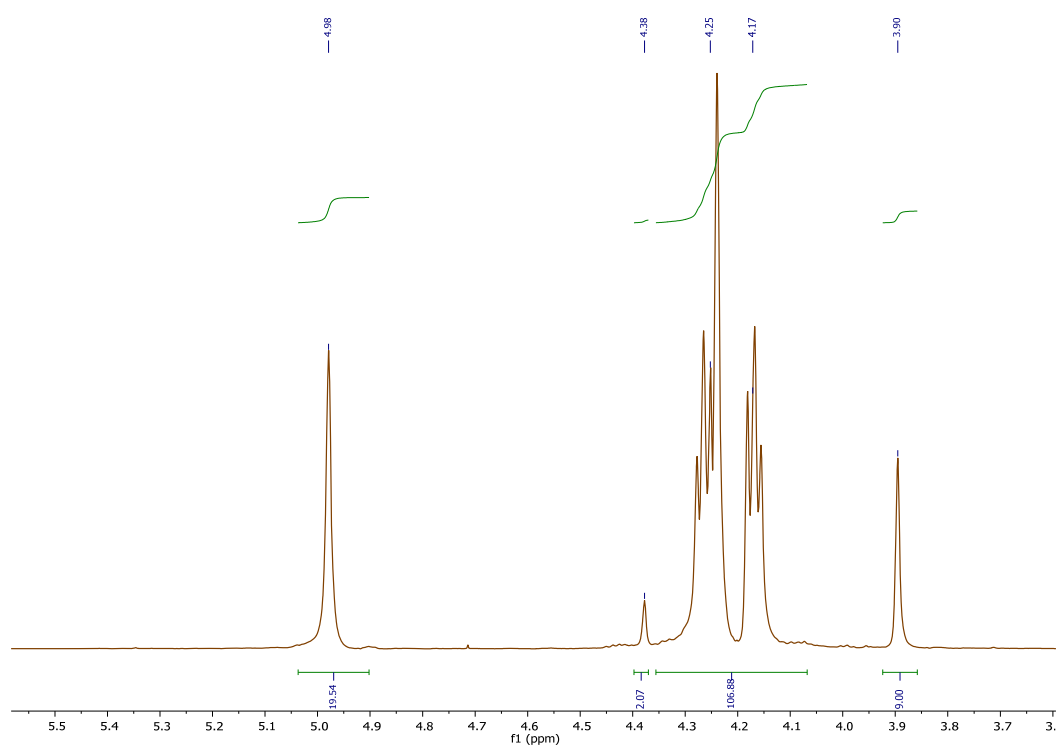
¹H-NMR (400 MHz): δ : 5.15 (s, OH, 4.21*2H), 4.37 (s, CH₂, 2H), 4.24-4.15 (m, CH₂-OH and CH₂-O-CH₂, 4.21*12H), 3.89 (s, NME₃, 9H)

Gb/D2 1:6.07



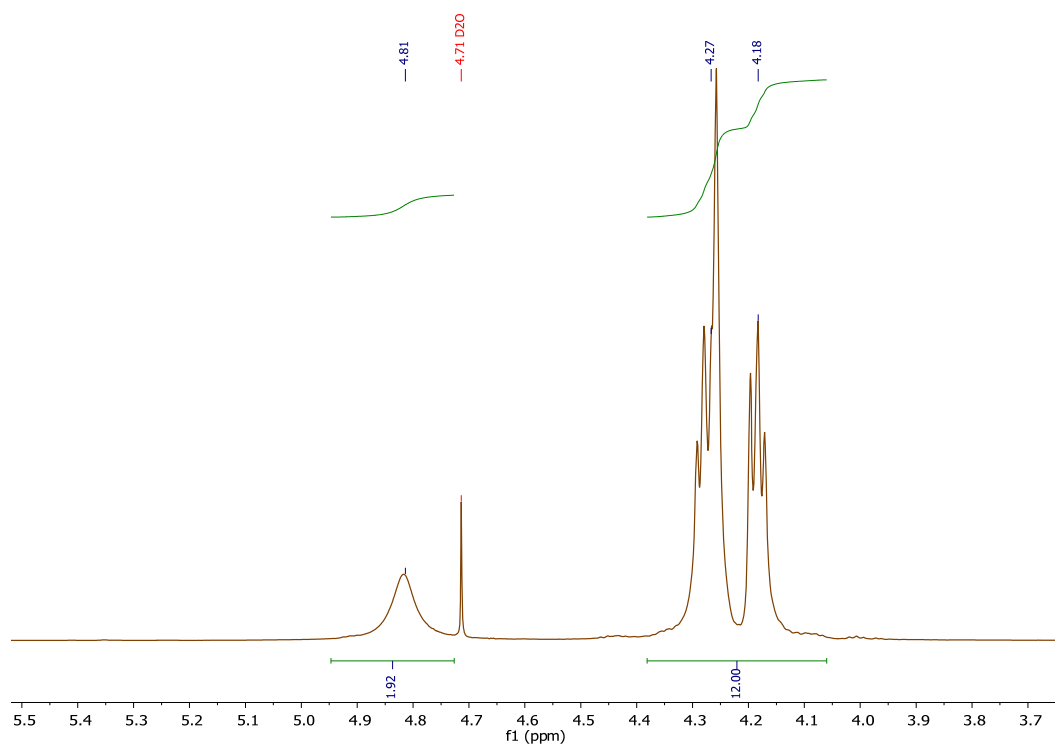
¹H-NMR (400 MHz): δ : 5.05 (s, OH, 6.07*2H), 4.38 (s, CH₂, 2H), 4.25-4.16 (m, CH₂-OH and CH₂-O-CH₂, 6.07*12H), 3.89 (s, NME₃, 9H)

Gb/D2 1:8.91



¹H-NMR (400 MHz): δ : 4.98 (s, OH, 8.91*2H), 4.38 (s, CH₂, 2H), 4.25-4.17 (m, CH₂-OH and CH₂-O-CH₂, 8.91*12H), 3.90 (s, NME₃, 9H)

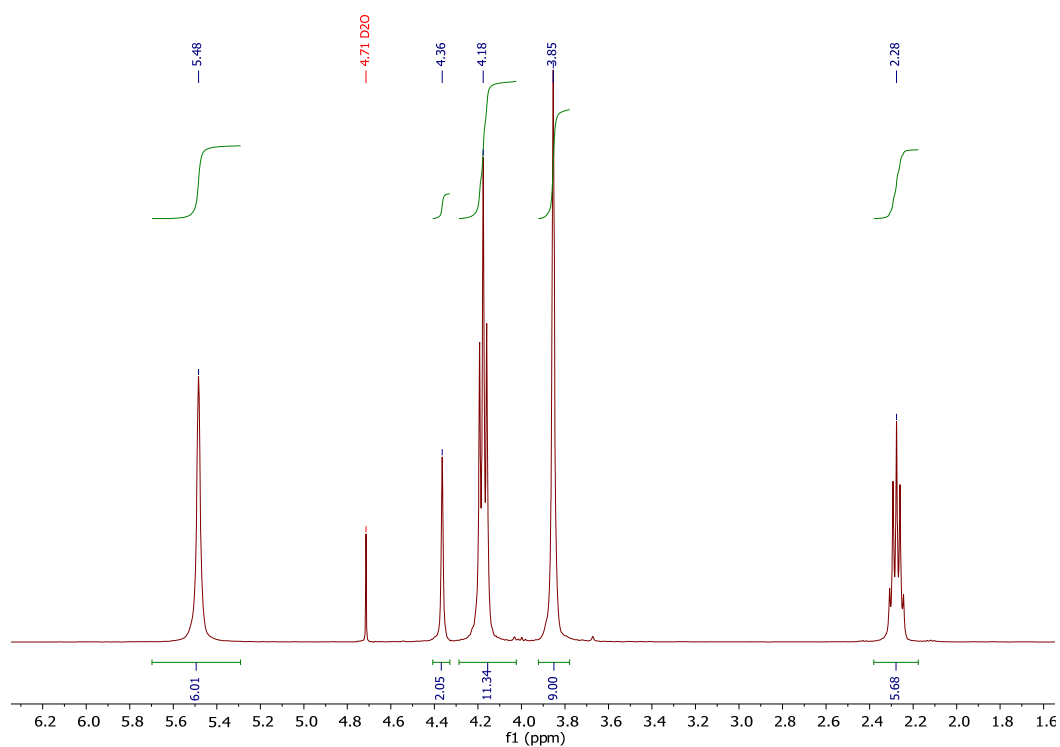
D2



¹H-NMR (400 MHz): δ : 4.81 (s, OH), 4.25-4.18 (m, CH₂-OH and CH₂-O-CH₂, 12H)

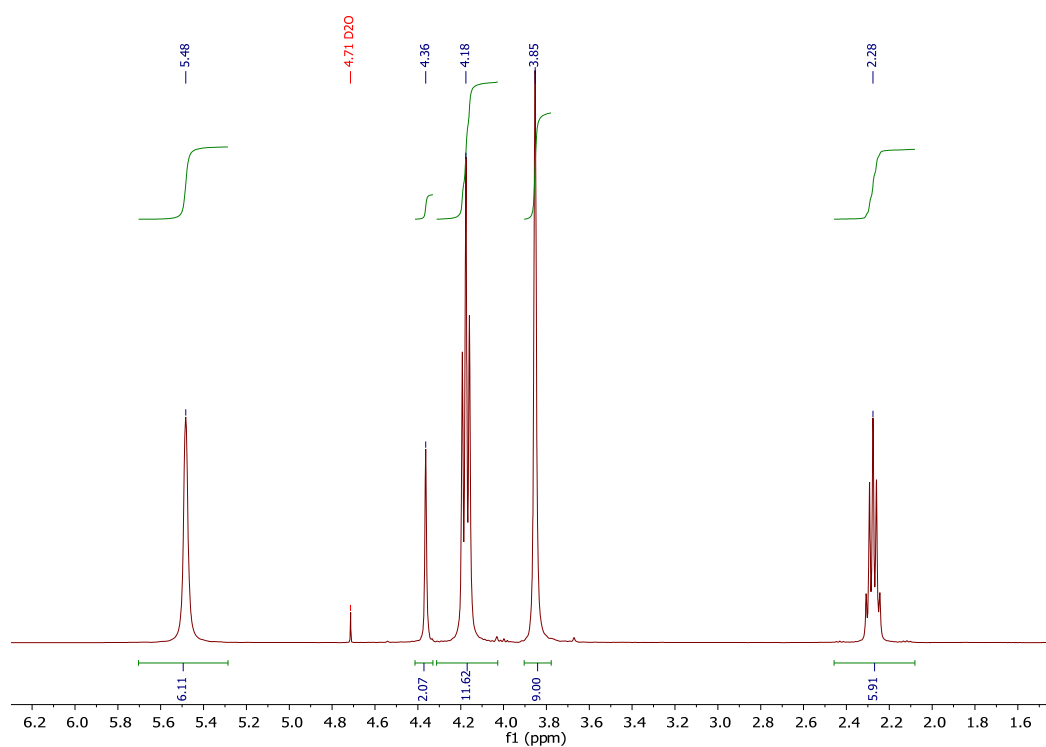
1.5. $^1\text{H-NMR}$ spectra of Gb/D3 mixtures

Gb/D3 1:2.86



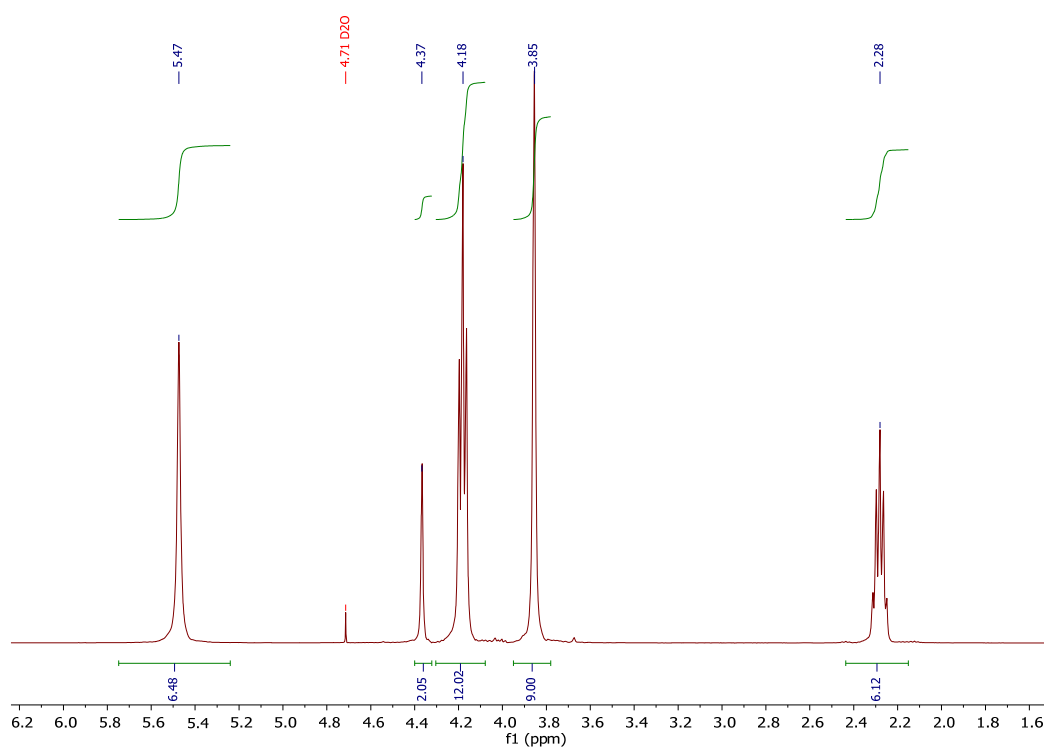
$^1\text{H-NMR}$ (400 MHz): δ : 5.48 (s, OH, 2.86*2H), 4.36 (s, CH_2 , 2H), 4.18 (t, $\text{CH}_2\text{-OH}$, 2.86*4H), 3.85 (s, NMe_3 , 9H), 2.28 (m, $\text{CH}_2\text{-CH}_2\text{-CH}_2$, 2.86*2H)

Gb/D3 1:2.93



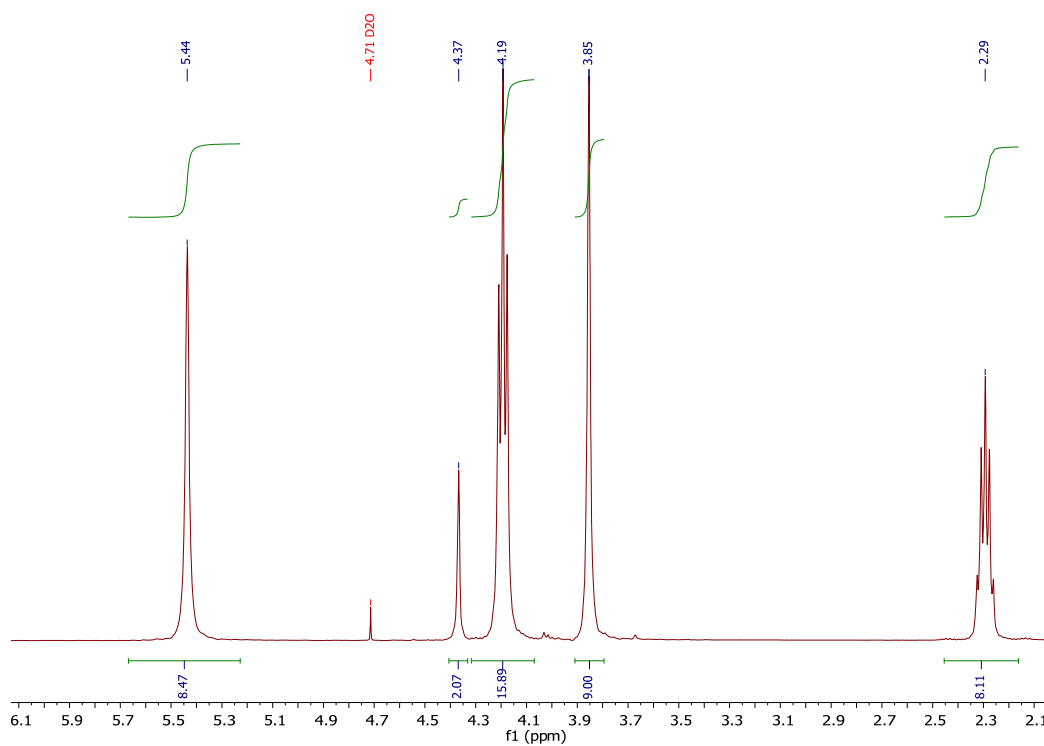
$^1\text{H-NMR}$ (400 MHz): δ : 5.48 (s, OH, 2.93*2H), 4.36 (s, CH_2 , 2H), 4.18 (t, $\text{CH}_2\text{-OH}$, 2.93*4H), 3.85 (s, NMe_3 , 9H), 2.28 (m, $\text{CH}_2\text{-CH}_2\text{-CH}_2$, 2.93*2H)

Gb/D3 1:3.10



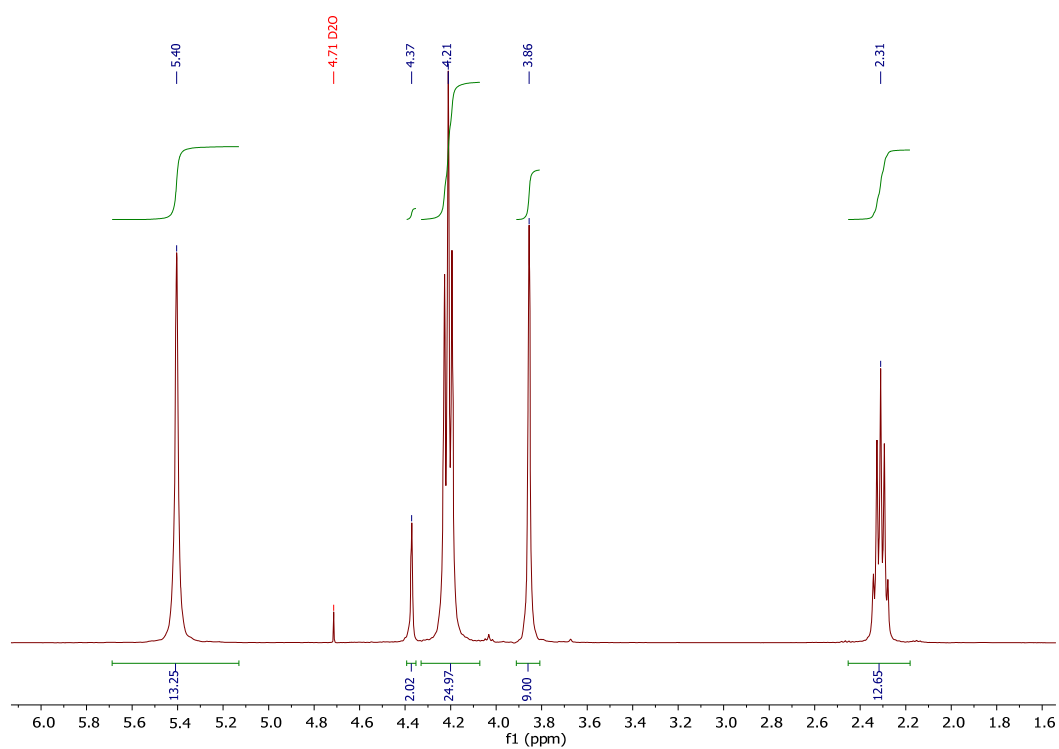
¹H-NMR (400 MHz): δ : 5.47 (s, OH, 3.10*2H), 4.37 (s, CH₂, 2H), 4.18 (t, CH₂-OH, 3.10*4H), 3.85 (s, NME₃, 9H), 2.28 (m, CH₂-CH₂-CH₂, 3.10*2H)

Gb/D3 1:4.05

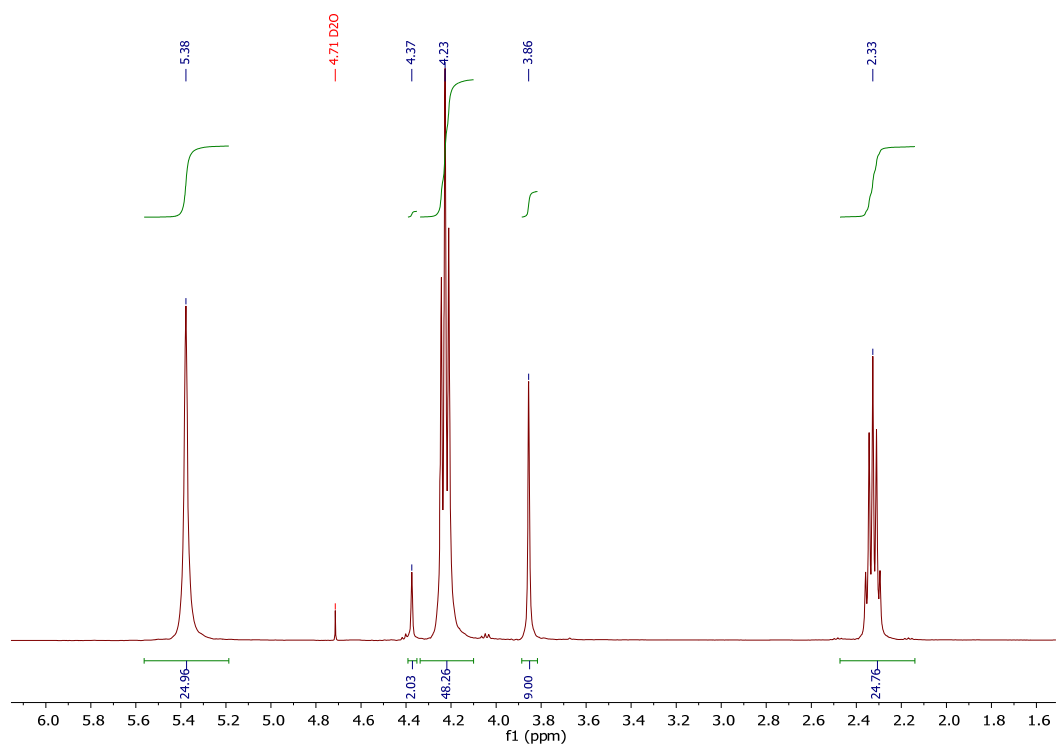


¹H-NMR (400 MHz): δ : 5.44 (s, OH, 4.05*2H), 4.37 (s, CH₂, 2H), 4.19 (t, CH₂-OH, 4.05*4H), 3.85 (s, NME₃, 9H), 2.29 (m, CH₂-CH₂-CH₂, 4.05*2H)

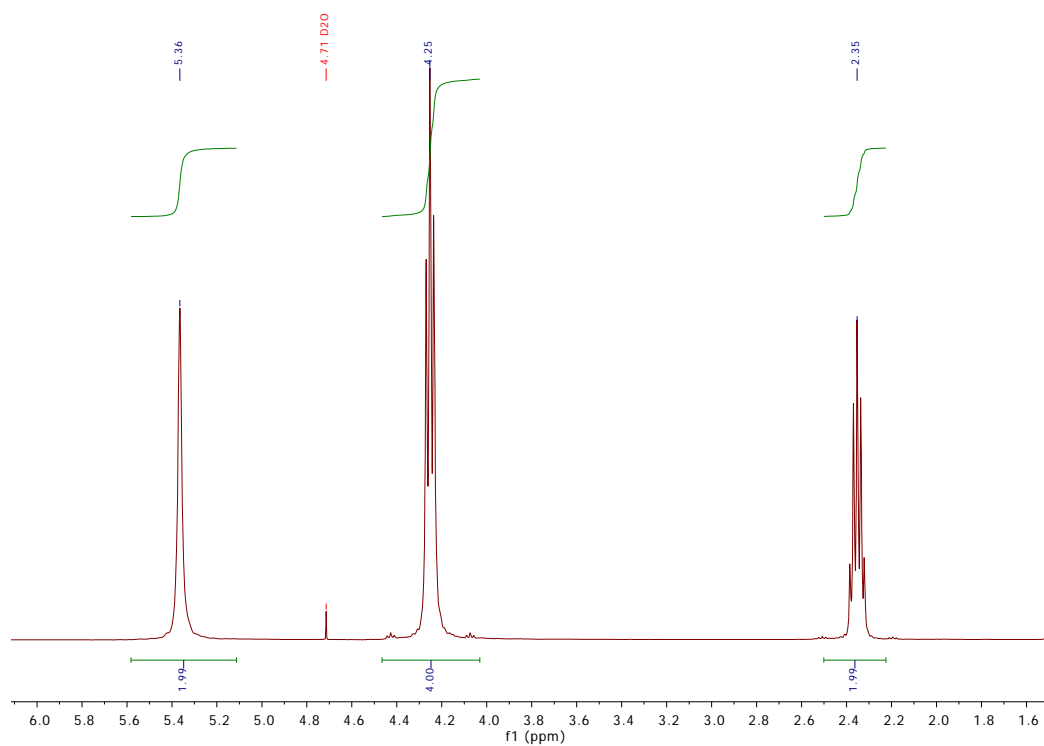
Gb/D3 1:6.32



Gb/D3 1:12.27



D3



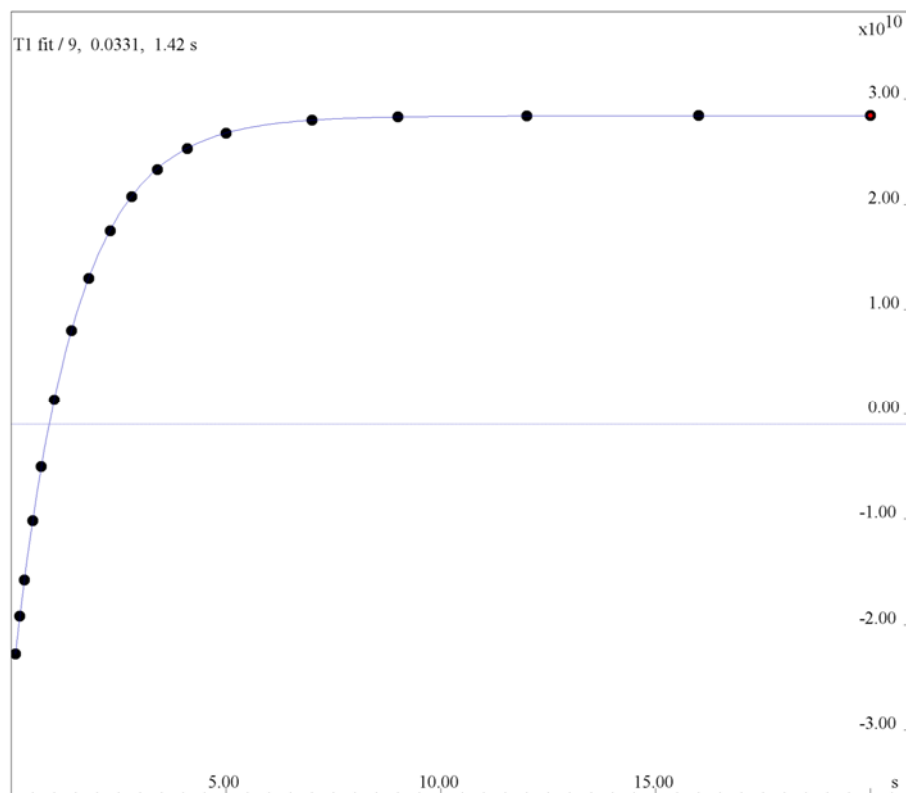
$^1\text{H-NMR}$ (400 MHz): δ : 5.36 (s, OH), 4.25 (t, $\text{CH}_2\text{-OH}$, 4H), 2.35 (m, $\text{CH}_2\text{-CH}_2\text{-CH}_2$, 2H)

2. Copies of T_1 exponential fitting by inversion recovery technique

2.1. T_1 exponential fitting of Gb/D1 mixtures

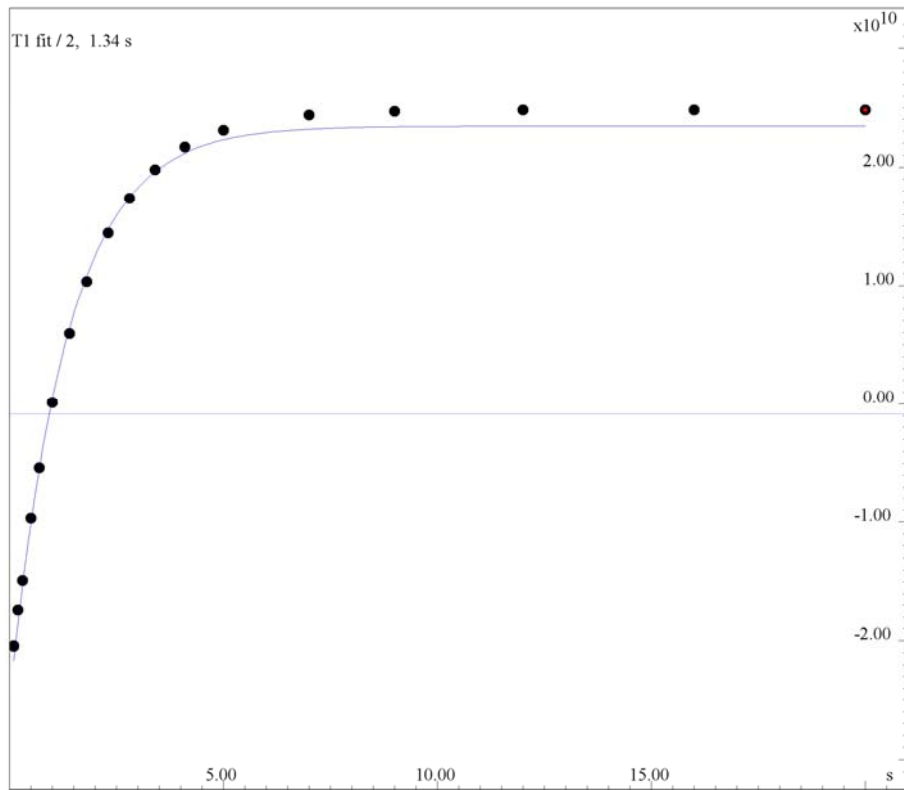
Gb/D1 1:2.44

T_1 : 1.42 sec \pm 1.244e-05

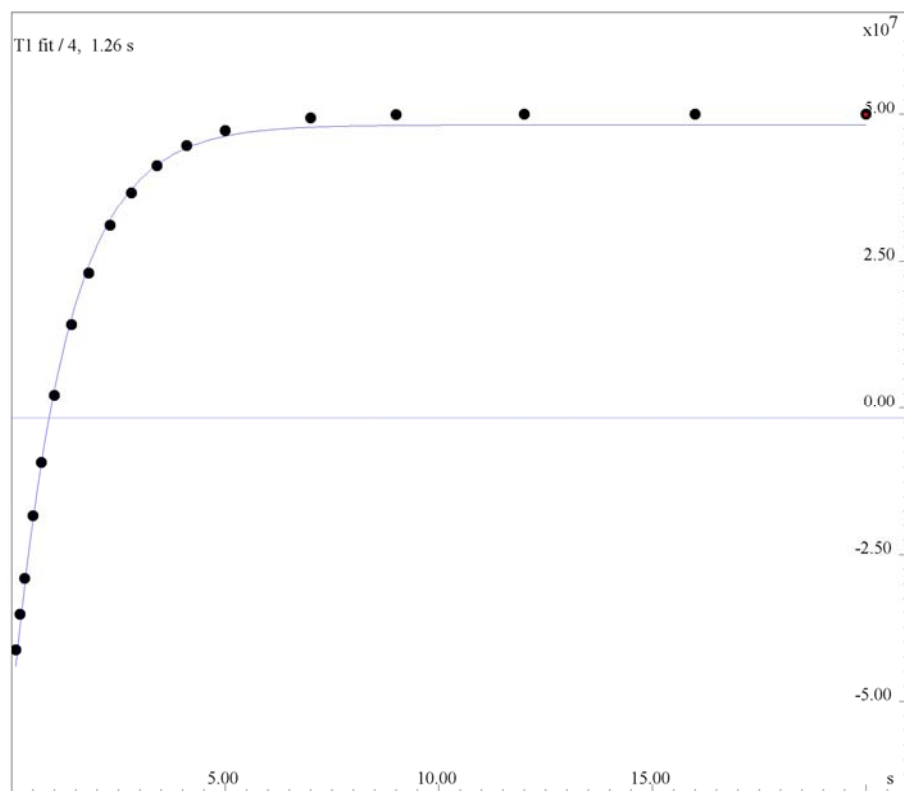


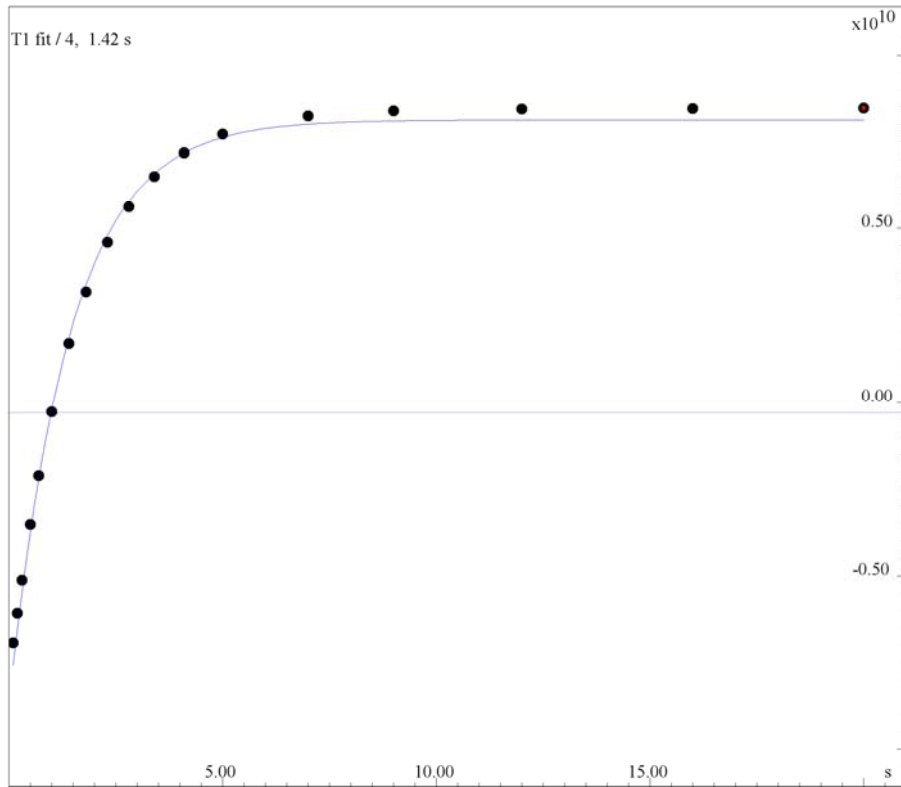
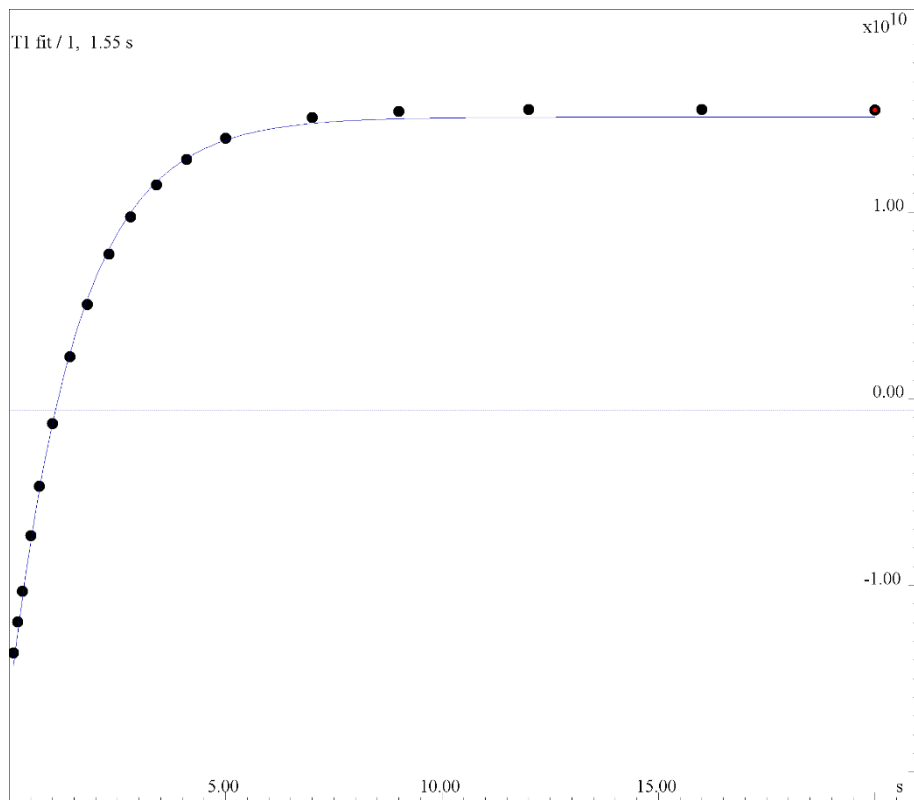
Gb/D1 1:2.73

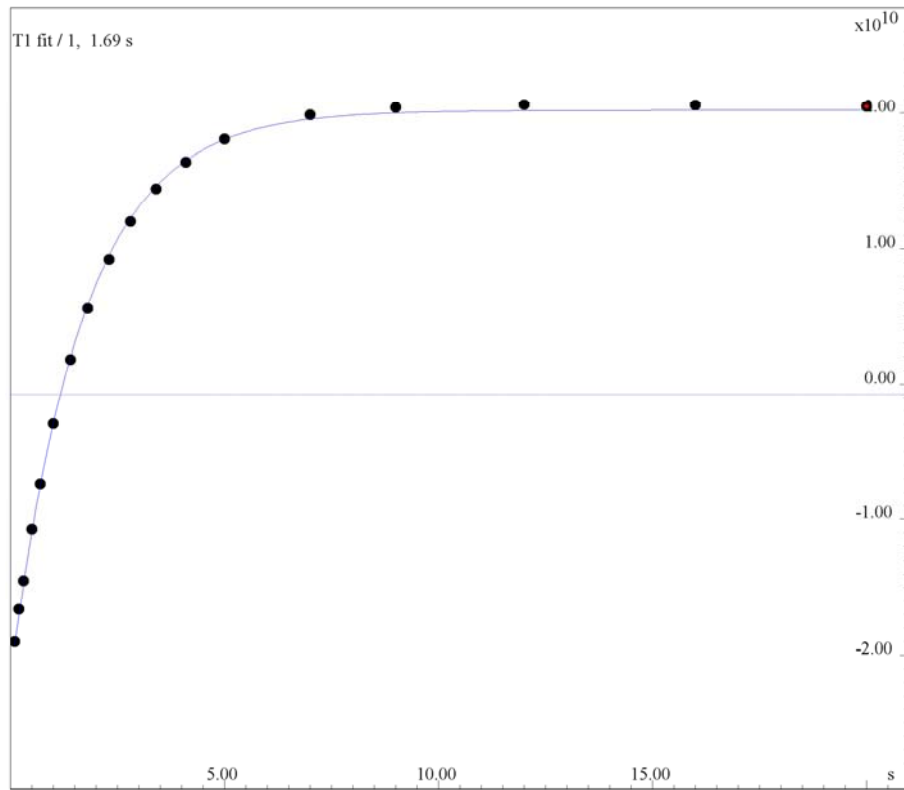
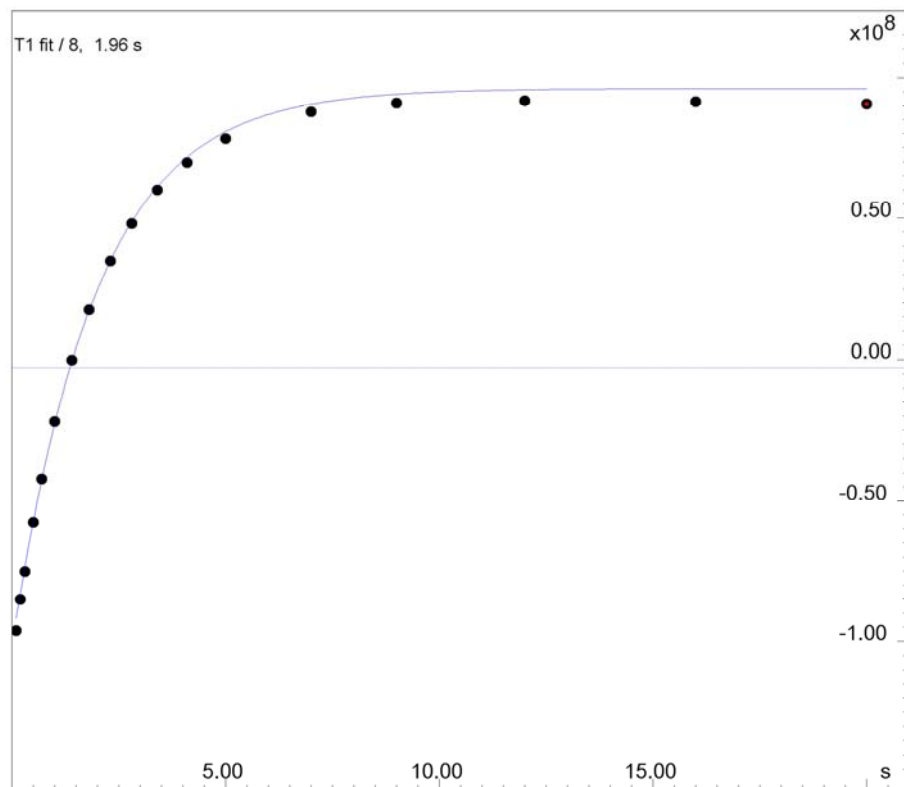
$$T_1: 1.34 \pm 2.188e-06$$

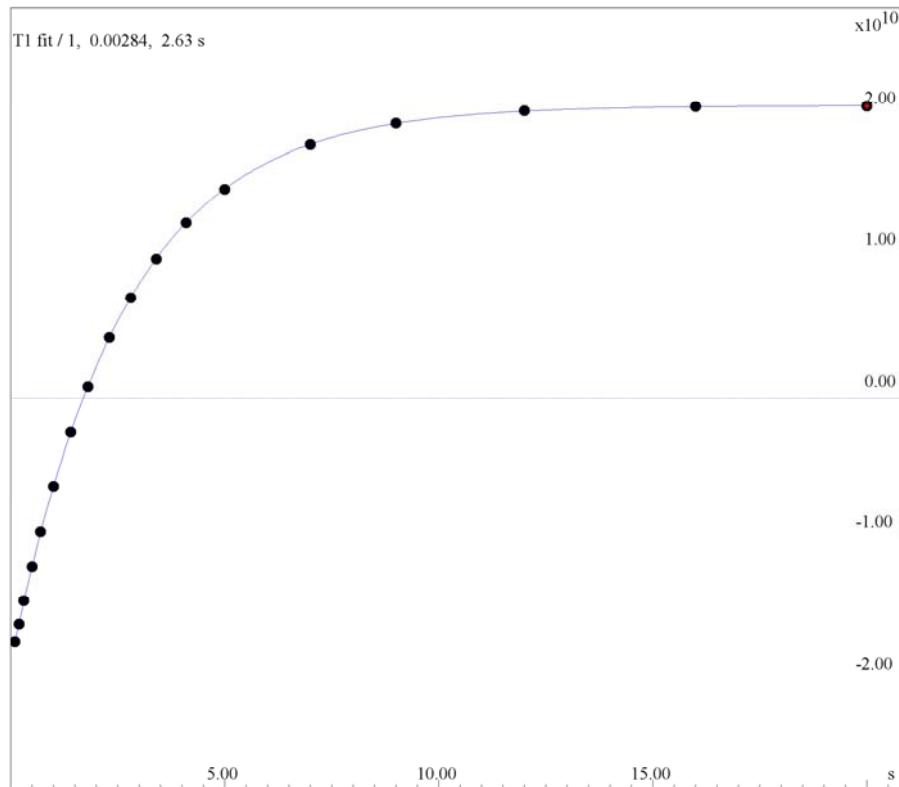
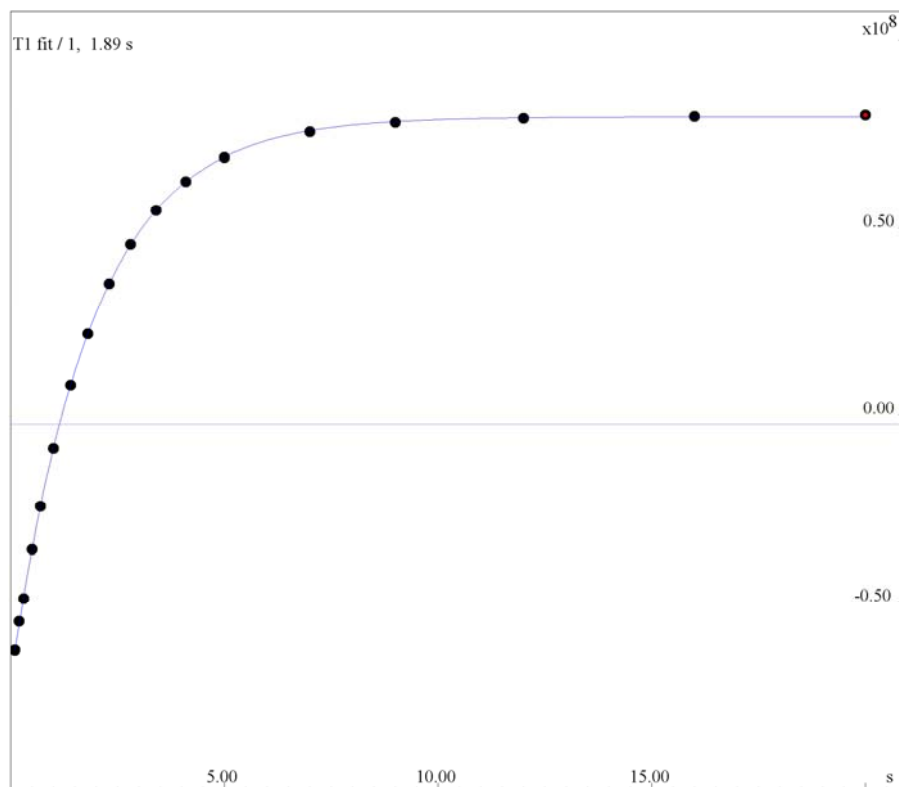
**Gb/D1 1:2.85**

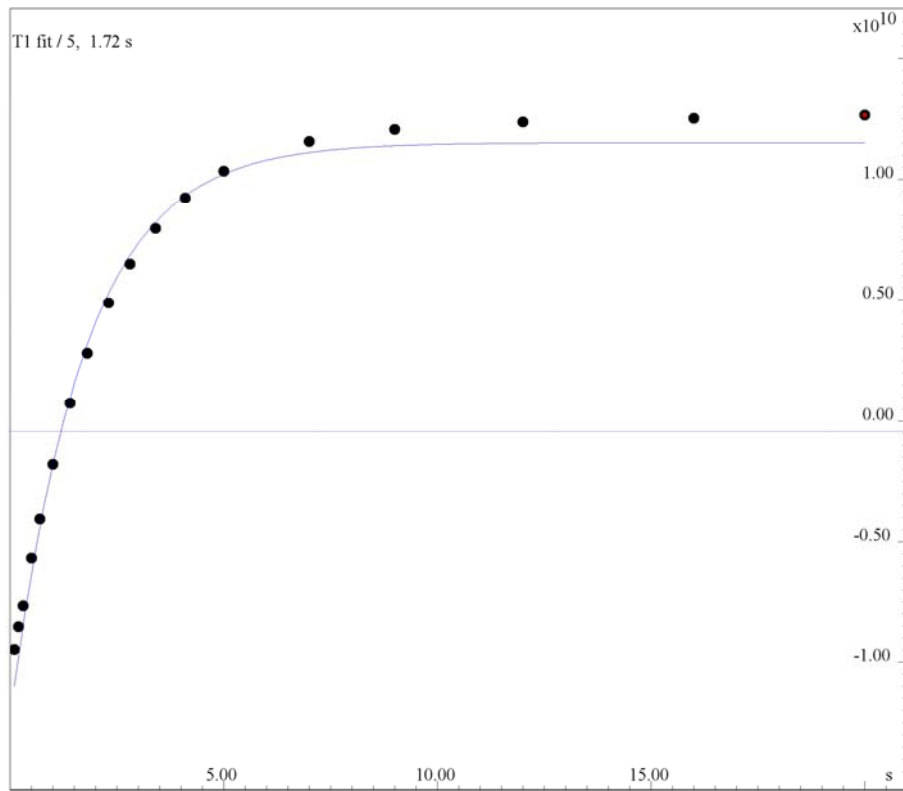
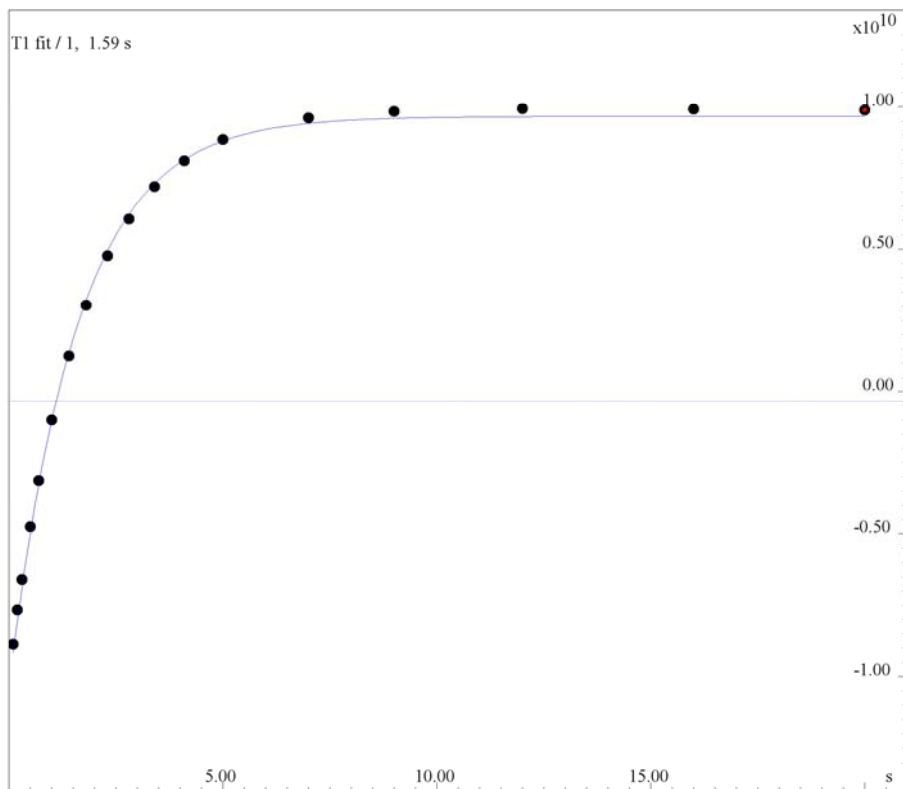
$$T_1: 1.26 \pm 2.585e-05$$

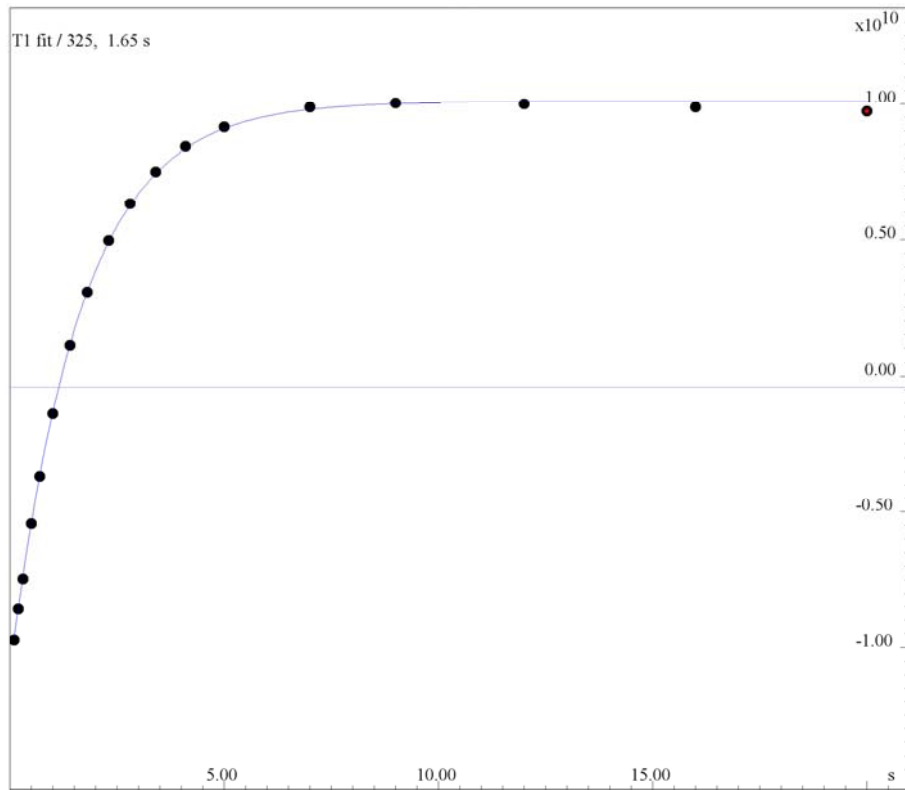
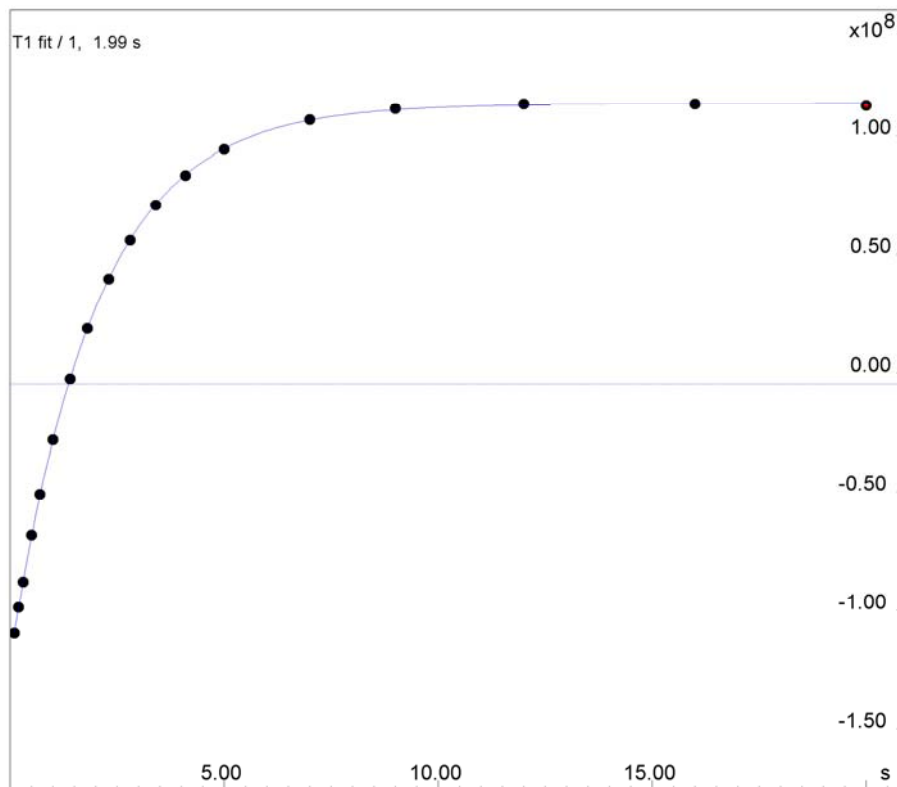


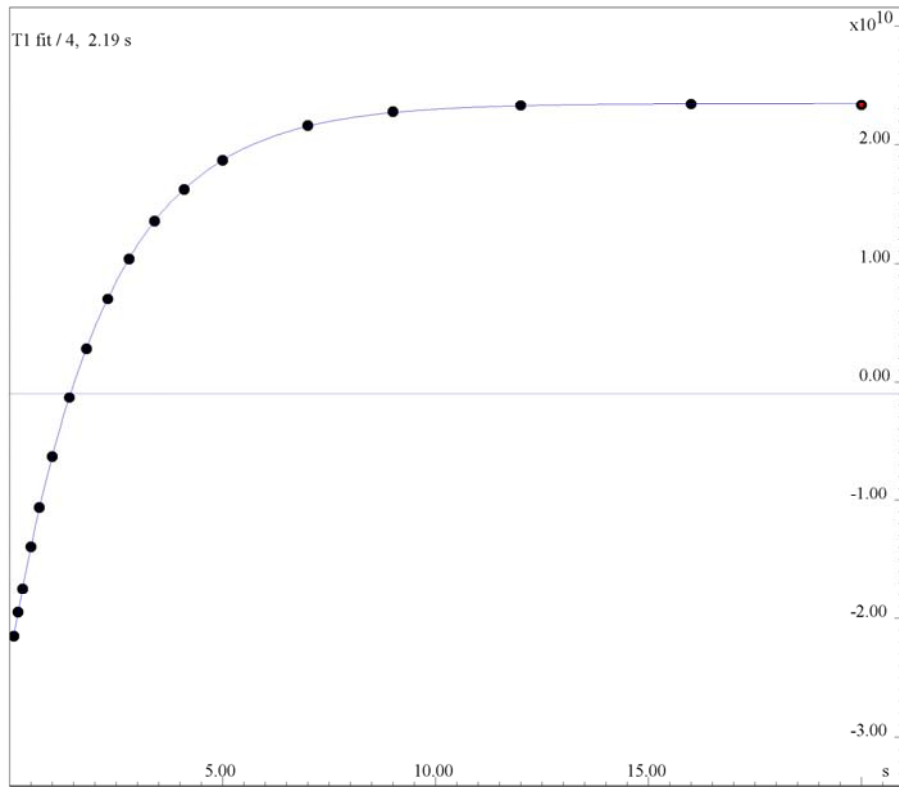
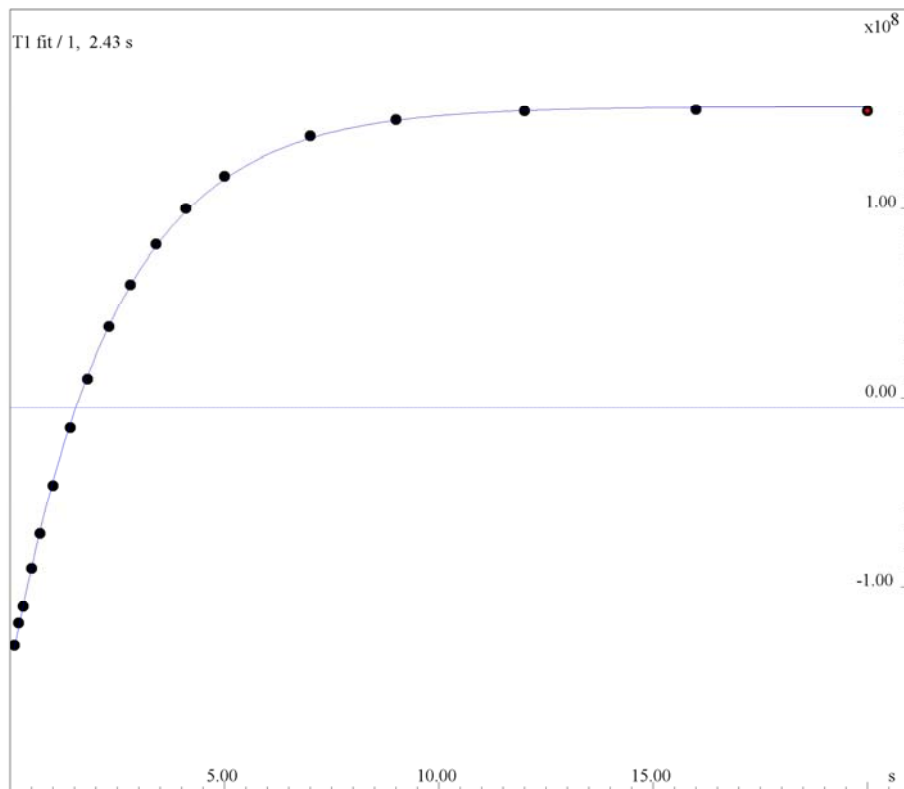
Gb/D1 1:3.18 $T_1: 1.42 \pm 0.059$ **Gb/D1 1:3.91** $T_1: 1.55 \pm 0.040$ 

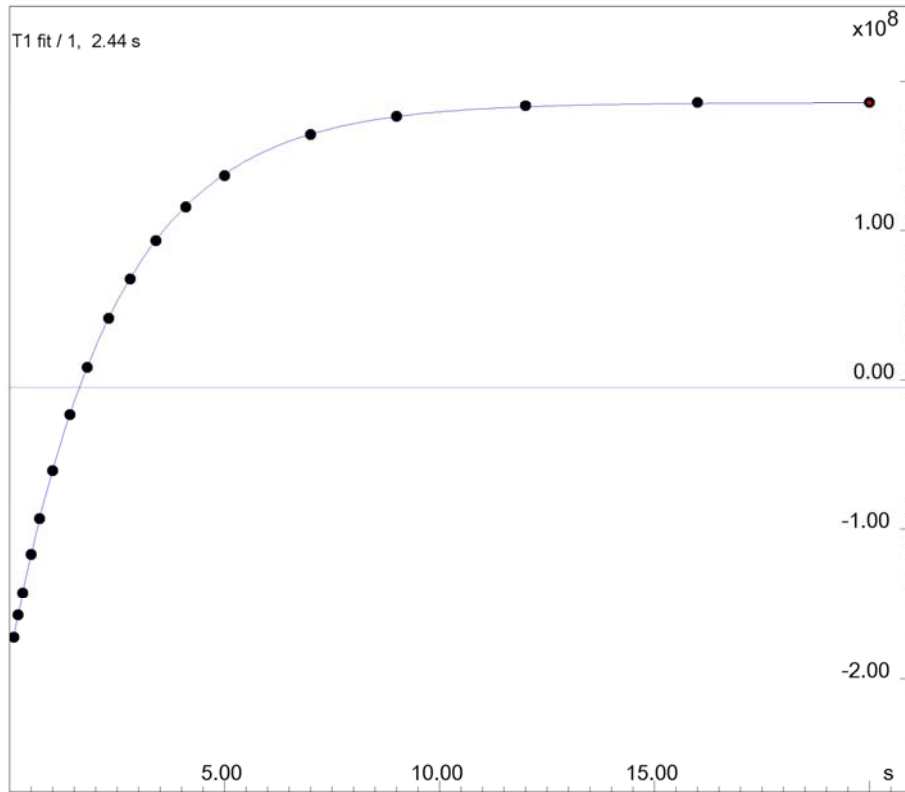
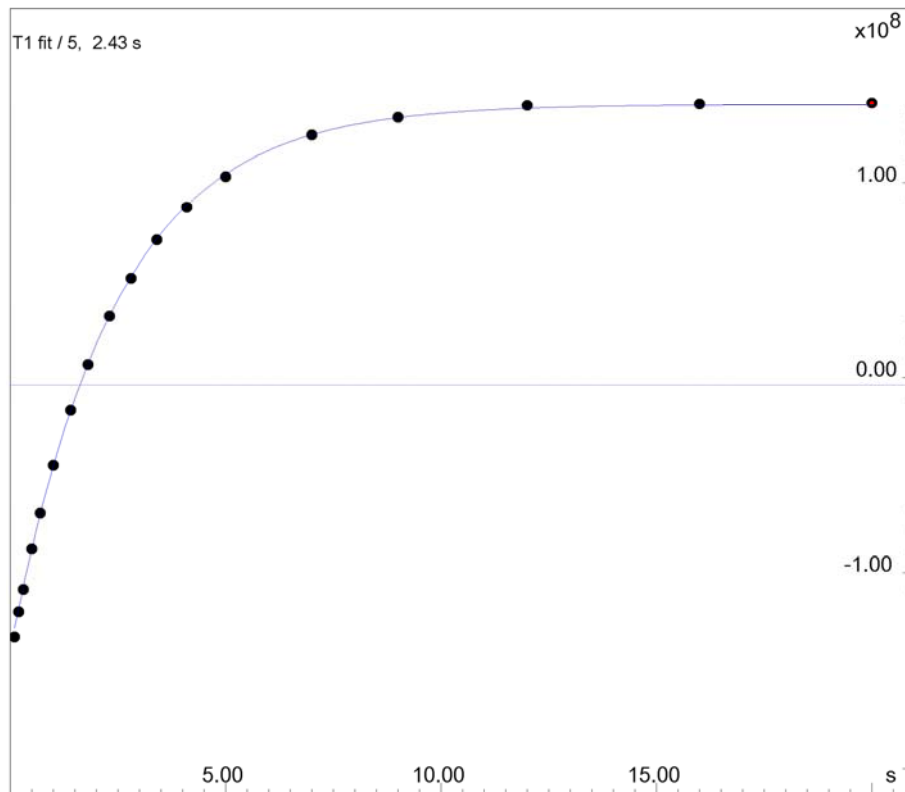
Gb/D1 1:4.85 $T_1: 1.69 \pm 2.675e-06$ **Gb/D1 1:8.65** $T_1: 1.96 \pm 1.964e-05$ 

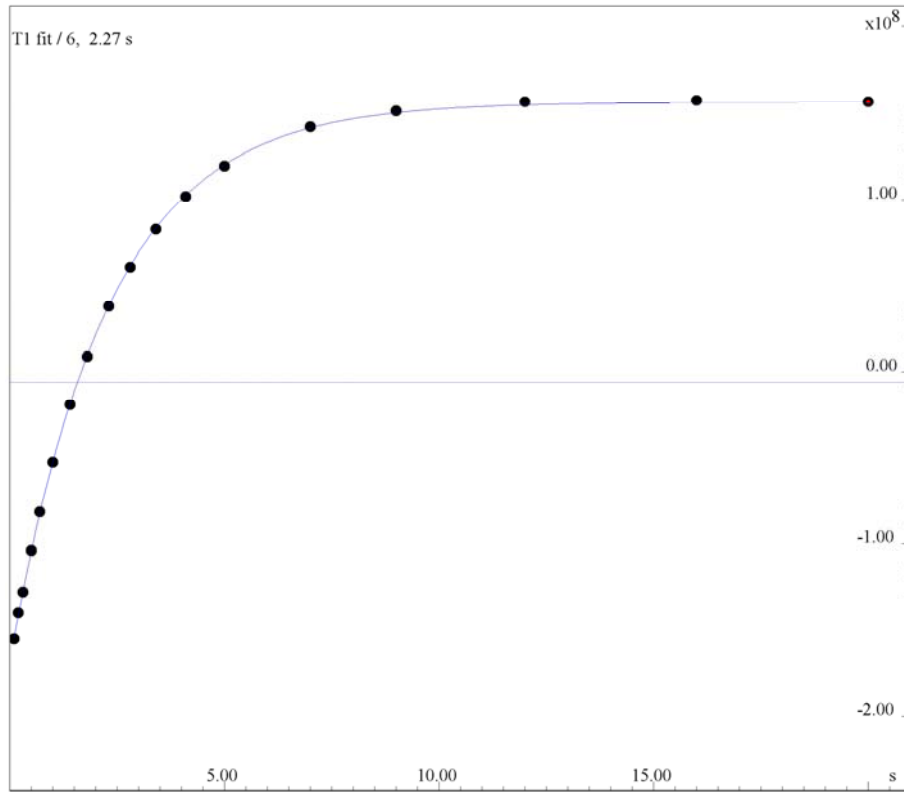
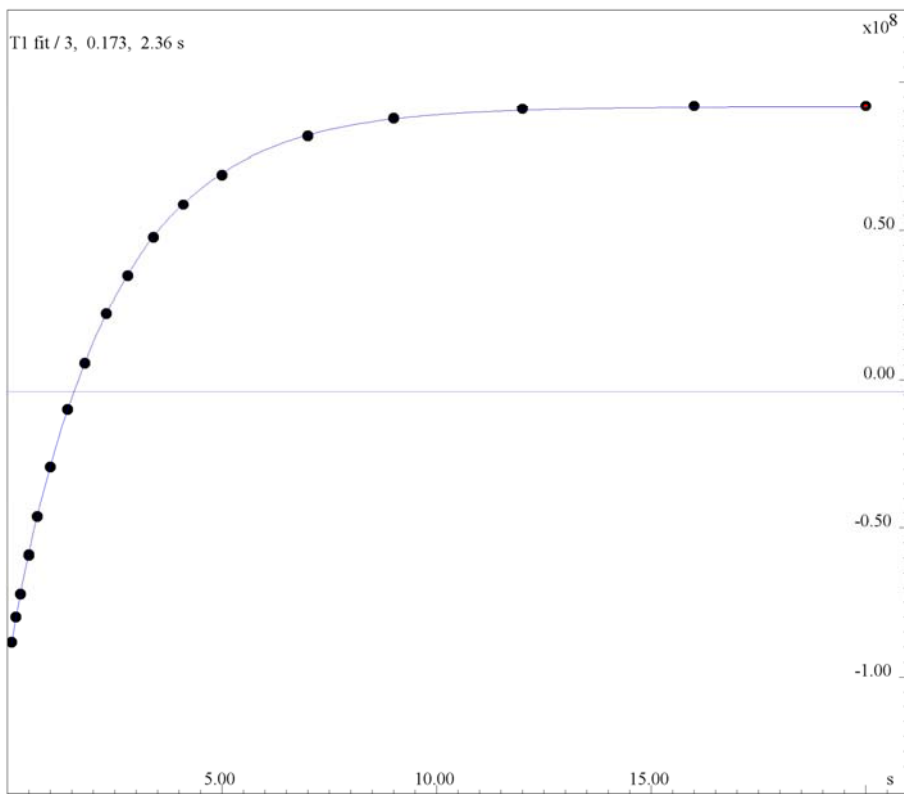
D1 $T_1: 2.63 \pm 4.870e-18$ **2.2. T_1 exponential fitting of Gb/D1 mixtures + 3 eq H₂O****Gb/D1 1:2.01 + 3 eq H₂O** $T_1: 1.89 \text{ sec} \pm 0.01278$ 

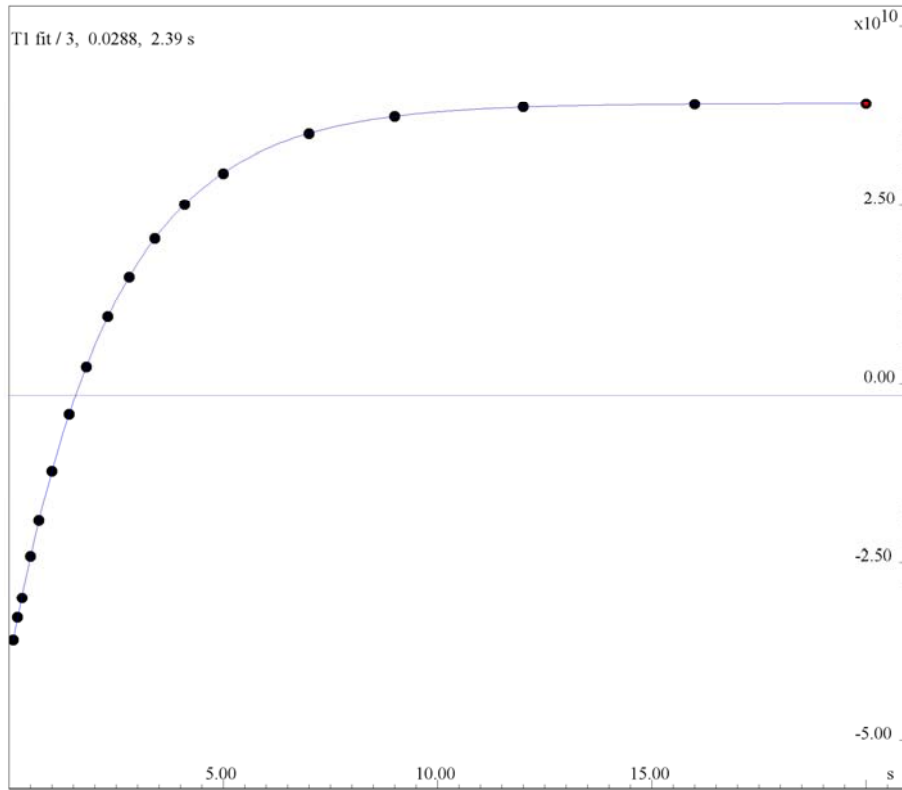
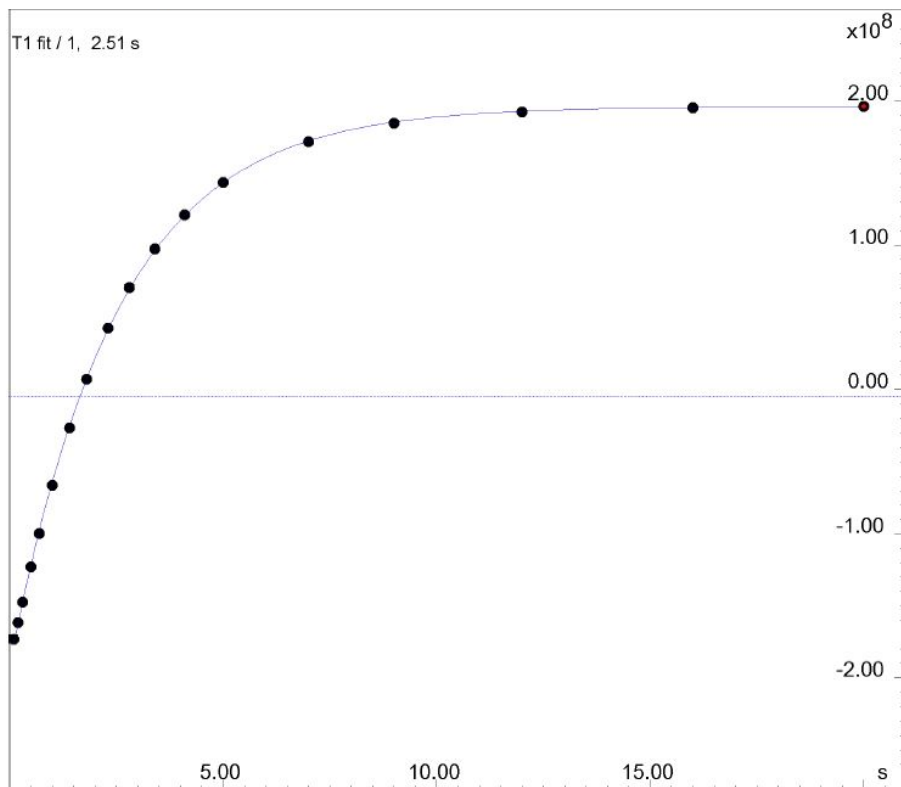
Gb/D1 1:2.49+3 eq H₂O $T_1: 1.72 \text{ sec} \pm 4.062\text{e-}06$ **Gb/D1 1:2.85+3 eq H₂O** $T_1: 1.59 \text{ sec} \pm 0.041$ 

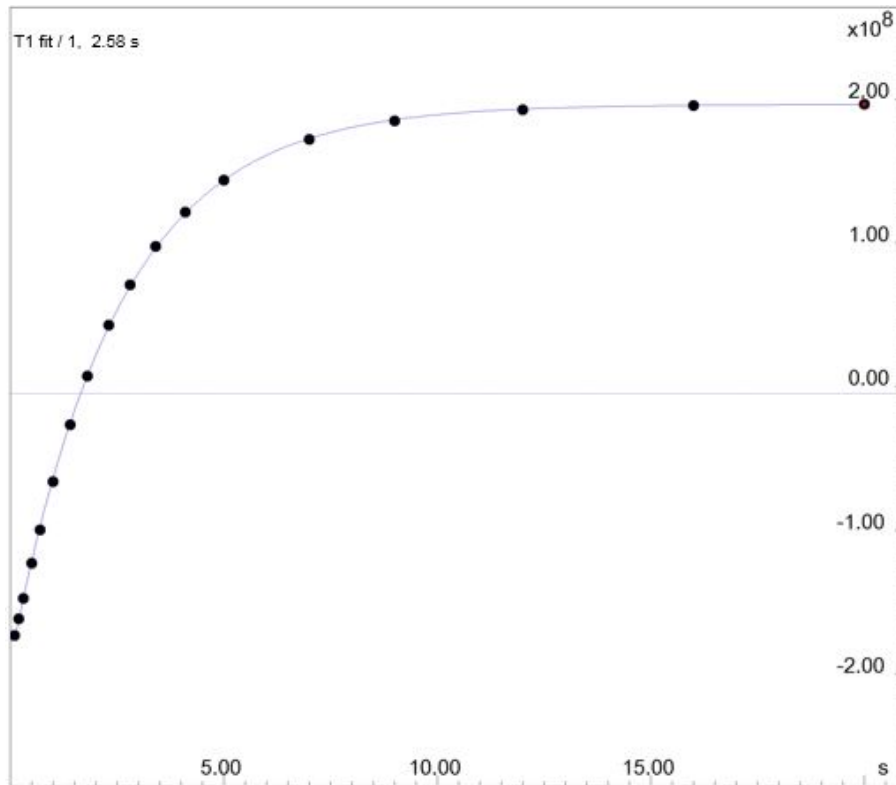
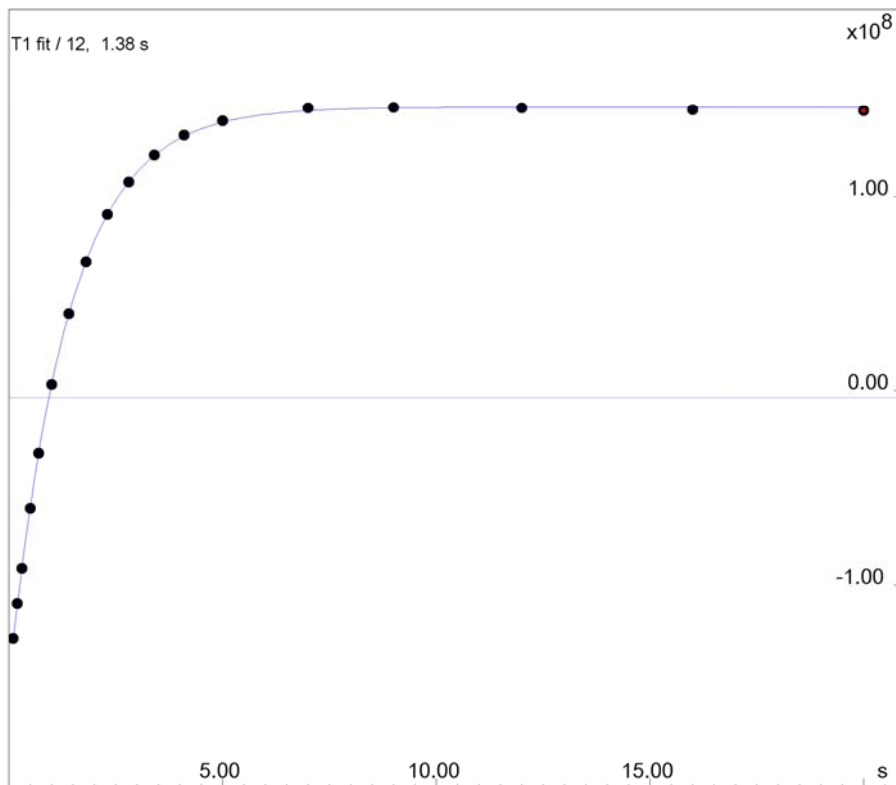
Gb/D1 1:3.11+3 eq H₂O $T_1: 1.65 \text{ sec} \pm 0.022$ **Gb/D1 1:5.76+3 eq H₂O** $T_1: 1.99 \text{ sec} \pm 0.011$ 

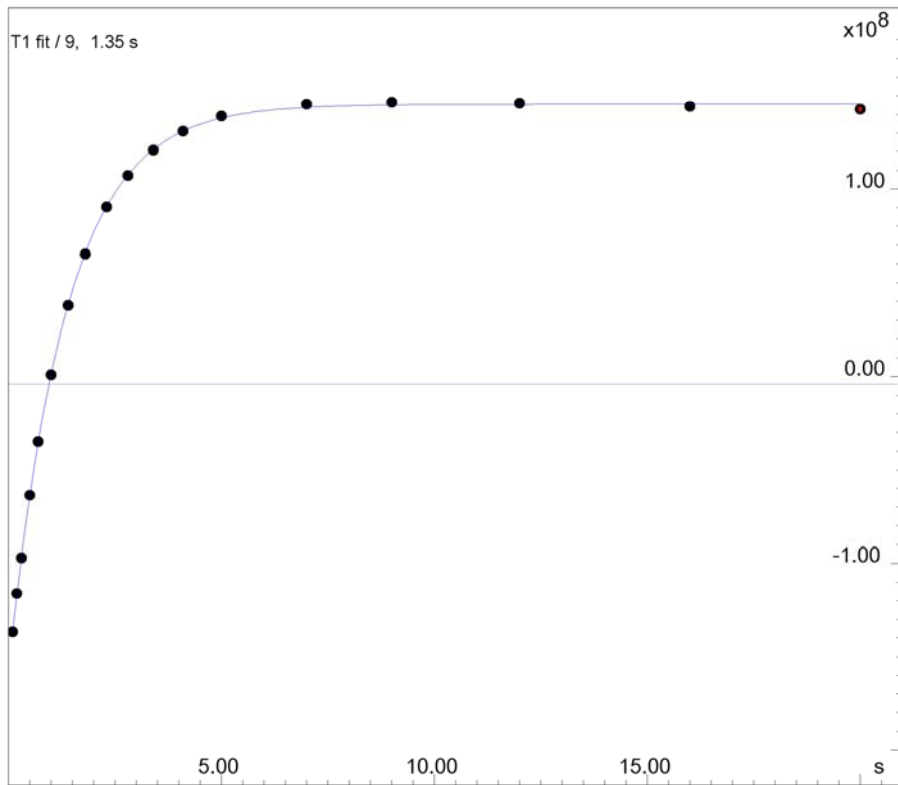
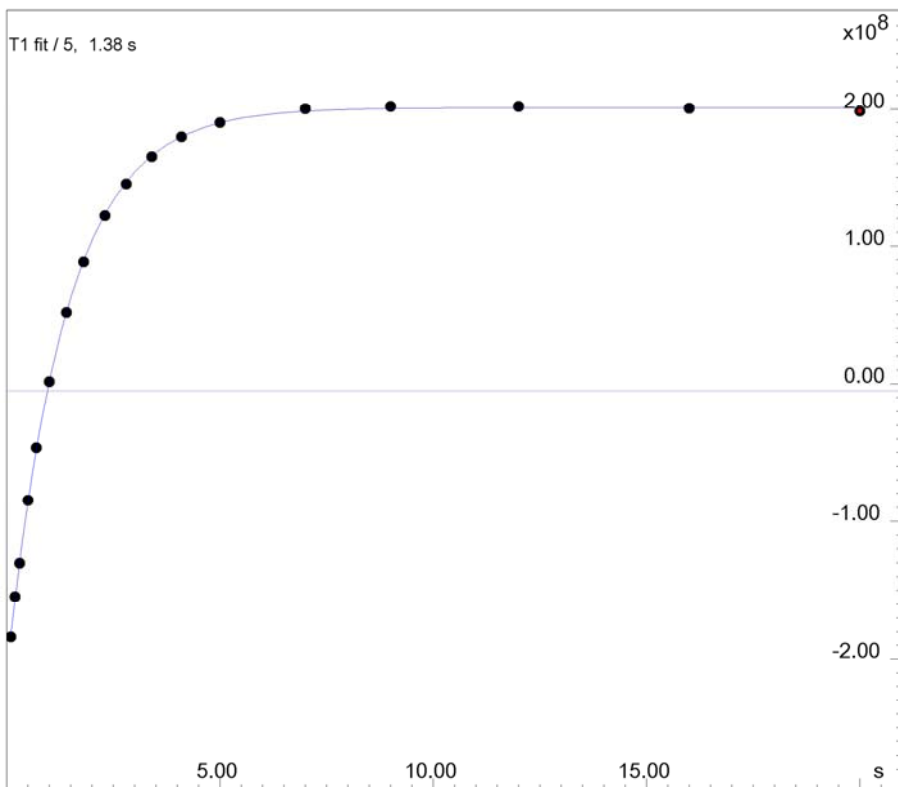
Gb/D1 1:11.81 + 3 eq H₂O $T_1: 2.19 \text{ sec} \pm 4.700\text{e-}06$ **Gb/D1 1:18.99 + 3 eq H₂O** $T_1: 2.43 \text{ sec} \pm 0.049$ 

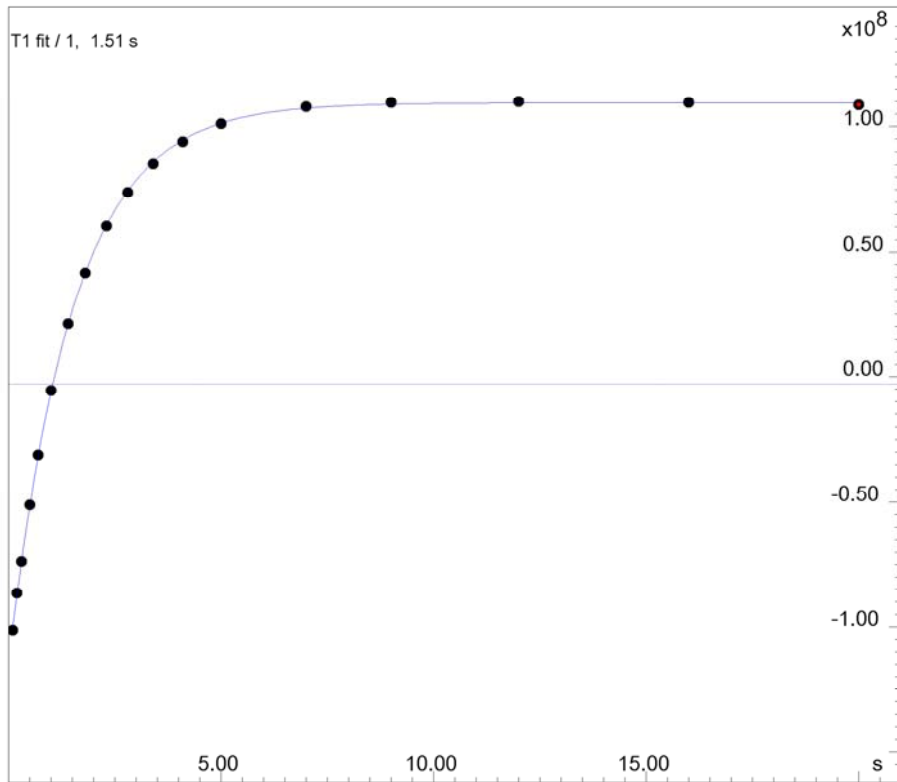
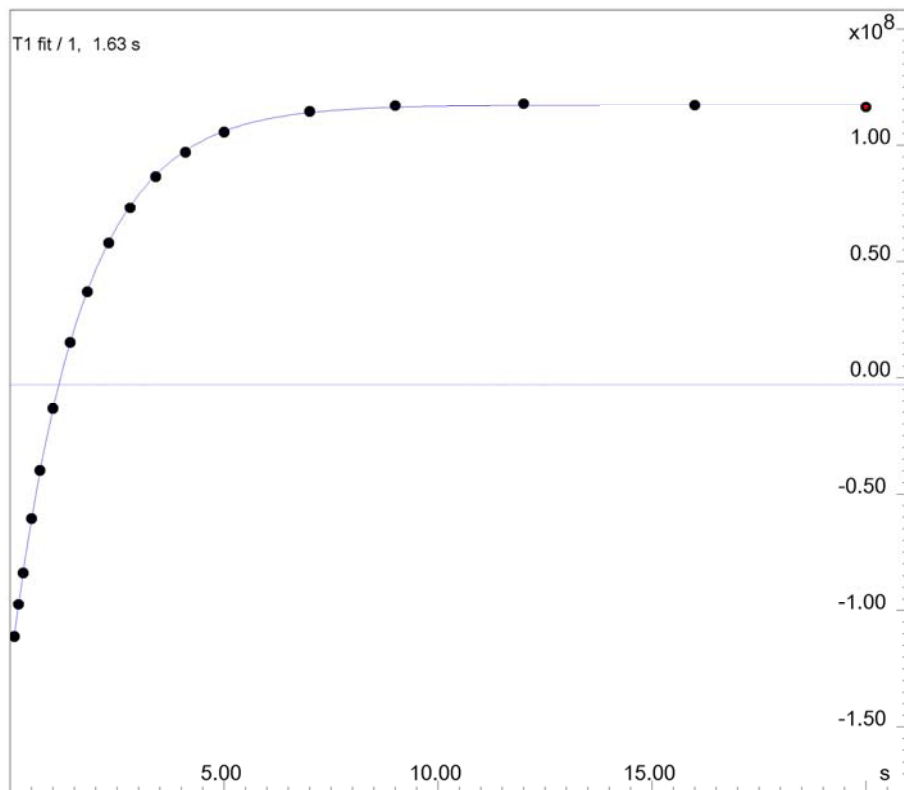
2.3. T_1 exponential fitting of Gb/D1 mixtures + 6 eq H_2O **Gb/D1 1:1.52 + 6 eq H_2O** T_1 : 2.44 sec \pm 6.288e-05**Gb/D1 1:2.02 + 6 eq H_2O** T_1 : 2.43 sec \pm 7.364e-05

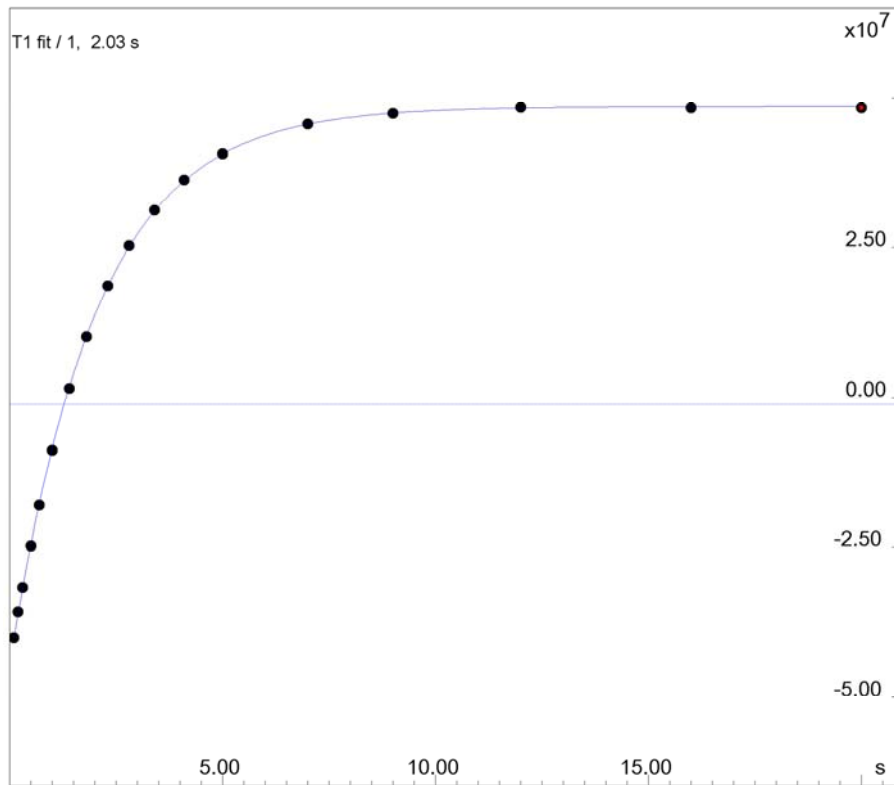
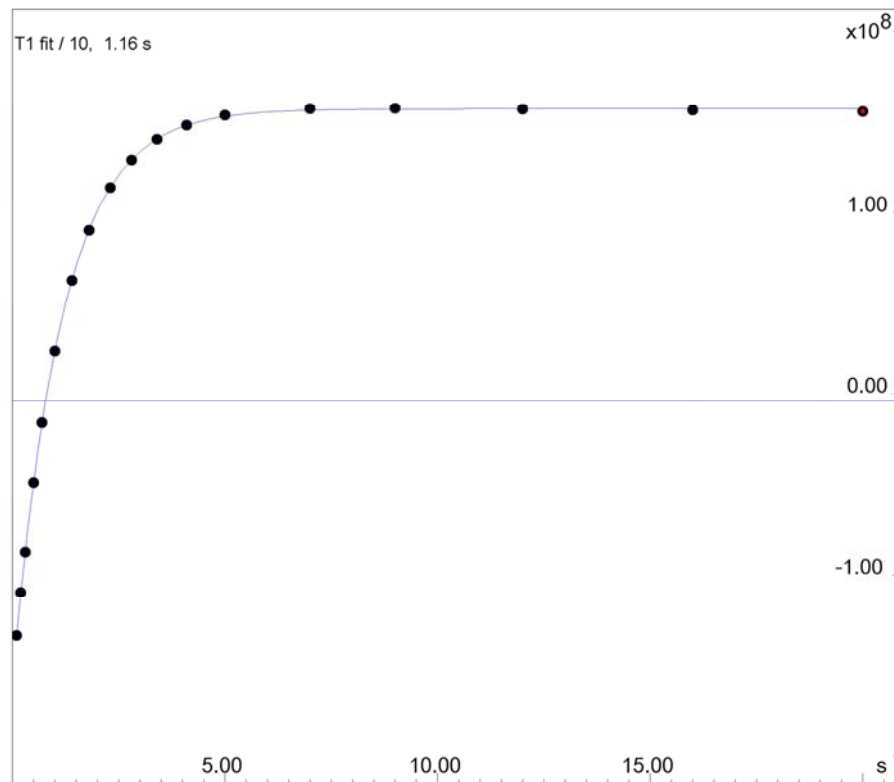
Gb/D1 1:3.02+6 eq H₂O $T_1: 2.27 \text{ sec} \pm 2.115\text{e-}05$ **Gb/D1 1:4.10+6 eq H₂O** $T_1: 2.36 \text{ sec} \pm 0.0001197$ 

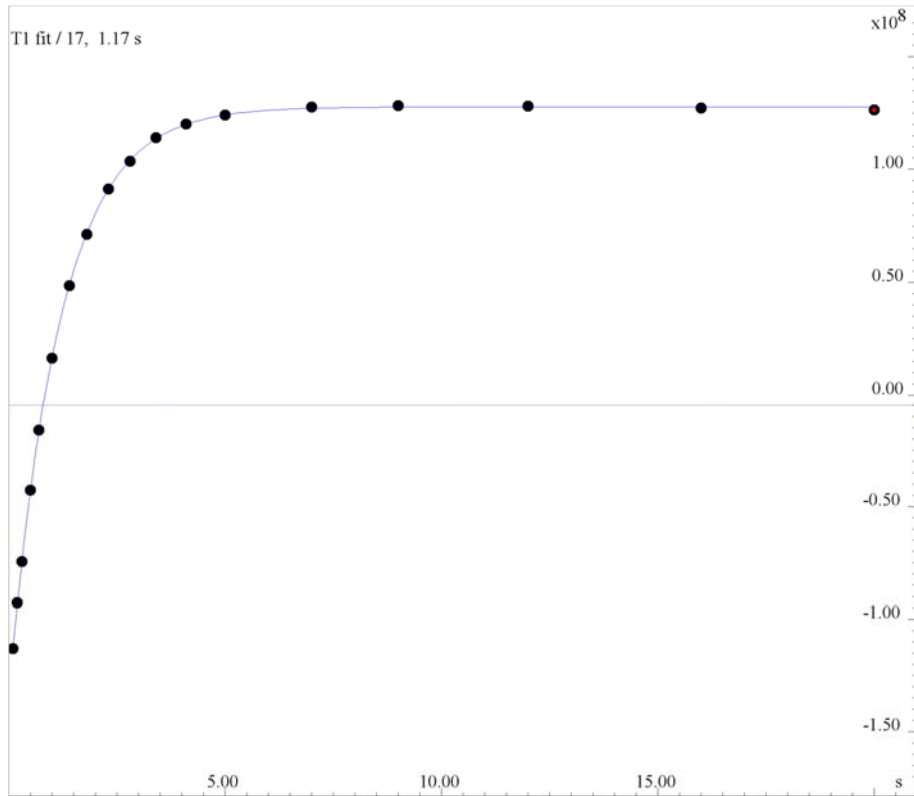
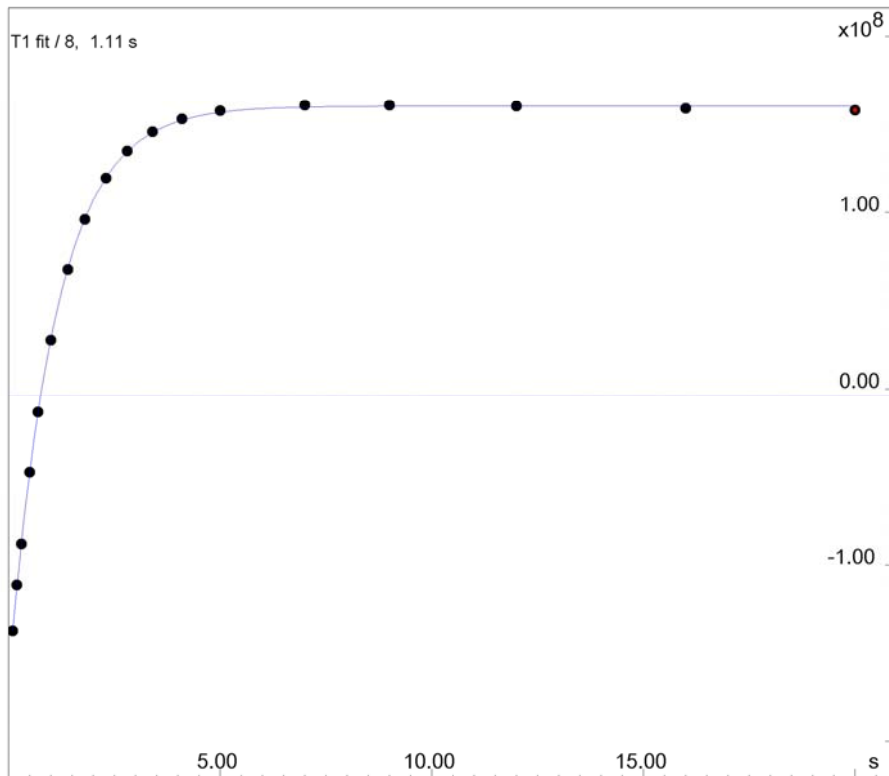
Gb/D1 1:6.05+6 eq H₂O $T_1: 2.39 \text{ sec} \pm 0.0099$ **Gb/D1 1:9.11+6 eq H₂O** $T_1: 2.51 \text{ sec} \pm 0.02683$ 

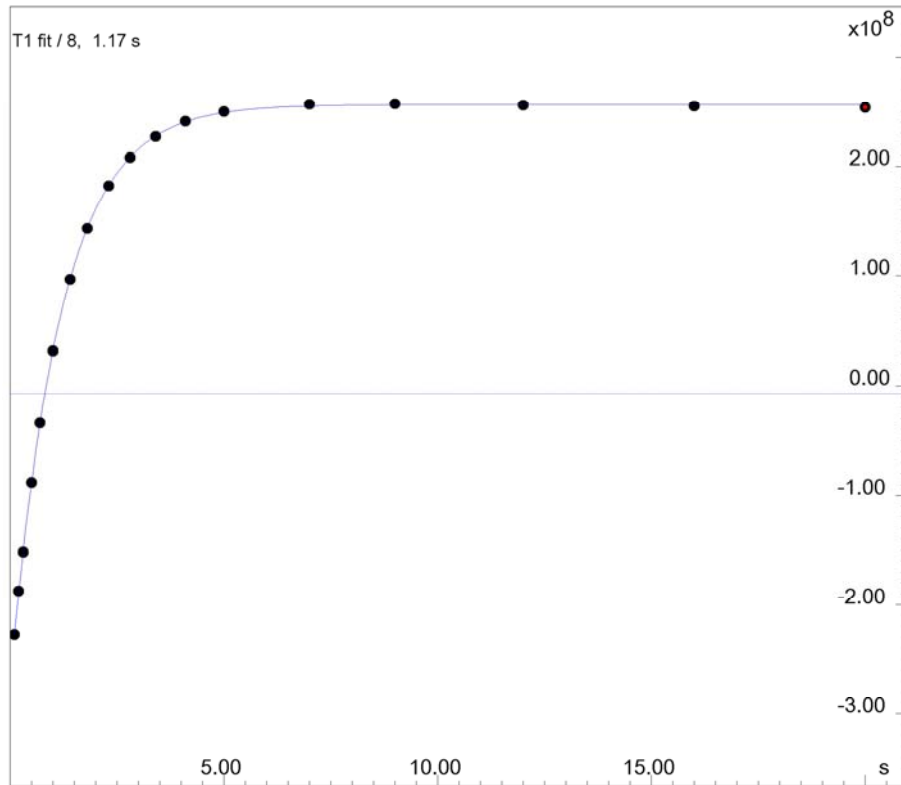
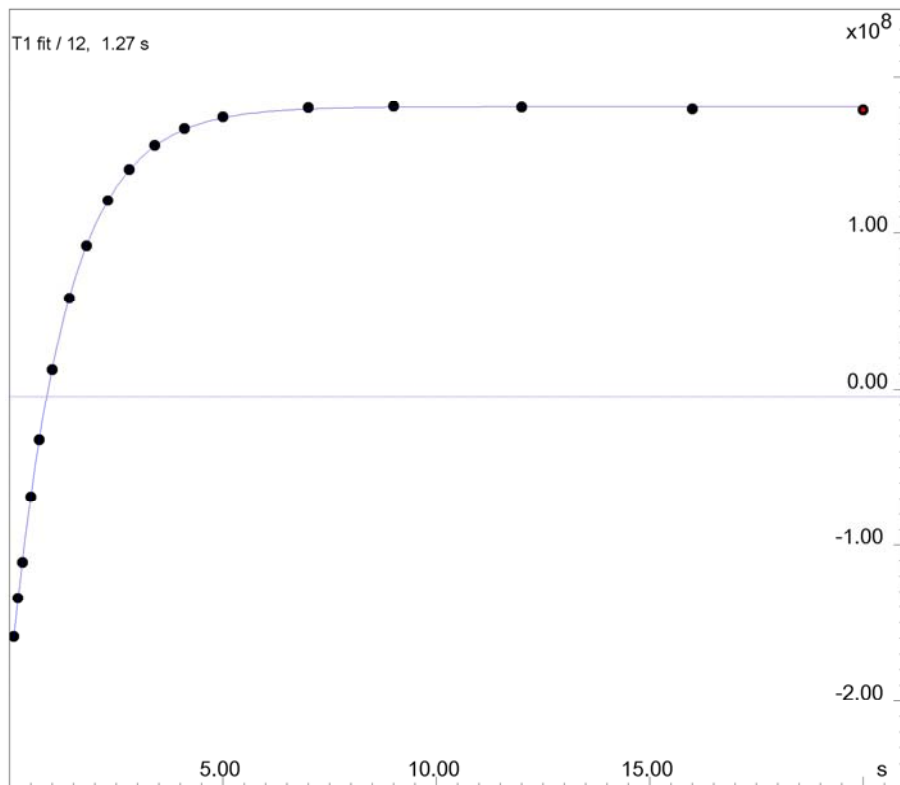
Gb/D1 1:11.82 + 6 eq H₂O $T_1: 2.58 \text{ sec} \pm 0.03752$ **2.4. T_1 exponential fitting of Gb/D2 mixtures****Gb/D2 1:3.66** $T_1: 1.38 \text{ sec} \pm 0.01627$ 

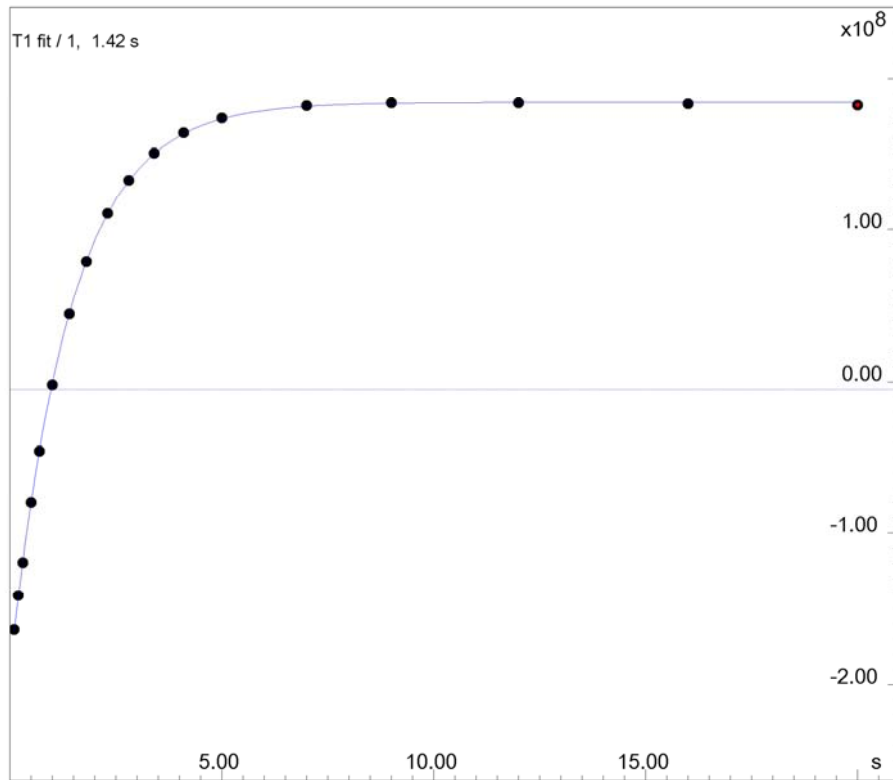
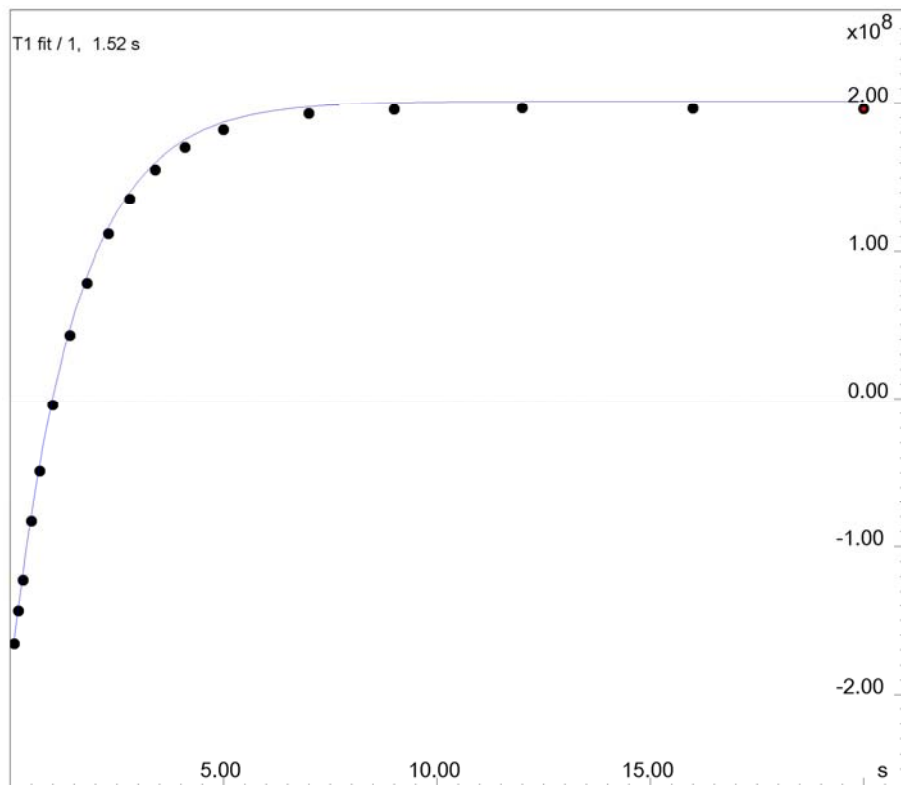
Gb/D2 1:3.76 $T_1: 1.35 \text{ sec} \pm 0.01837$ **Gb/D2 1:4.21** $T_1: 1.38 \text{ sec} \pm 0.01680$ 

Gb/D2 1:6.07 $T_1: 1.51 \text{ sec} \pm 0.01527$ **Gb/D2 1:8.91** $T_1: 1.63 \text{ sec} \pm 0.01547$ 

D2 $T_1: 2.03 \text{ sec} \pm 0.01993$ **2.5. T_1 exponential fitting of Gb/D3 mixtures****Gb/D3 1:2.86** $T_1: 1.16 \text{ sec} \pm 0.007736$ 

Gb/D3 1:2.93 $T_1: 1.17 \text{ sec} \pm 0.009845$ **Gb/D3 1:3.10** $T_1: 1.11 \text{ sec} \pm 0.01163$ 

Gb/D3 1:4.05 $T_1: 1.17 \text{ sec} \pm 0.008527$ **Gb/D3 1:6.32** $T_1: 1.27 \text{ sec} \pm 0.01324$ 

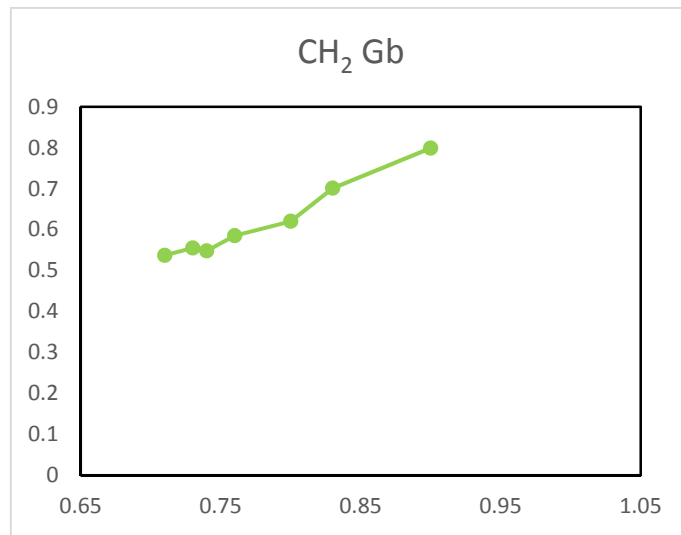
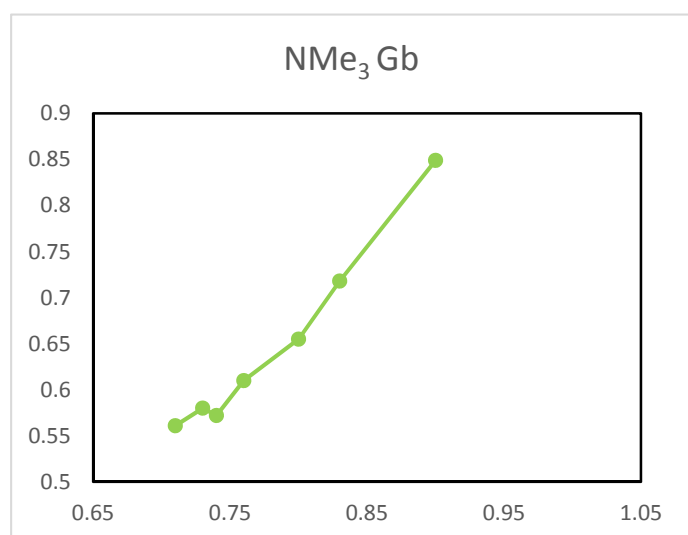
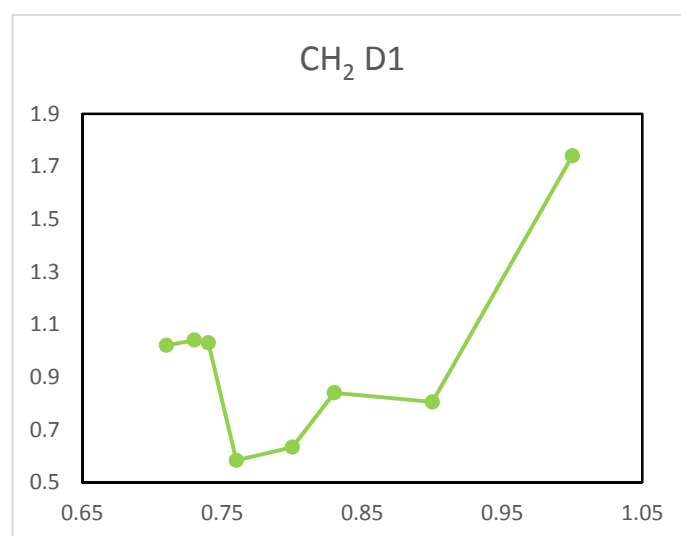
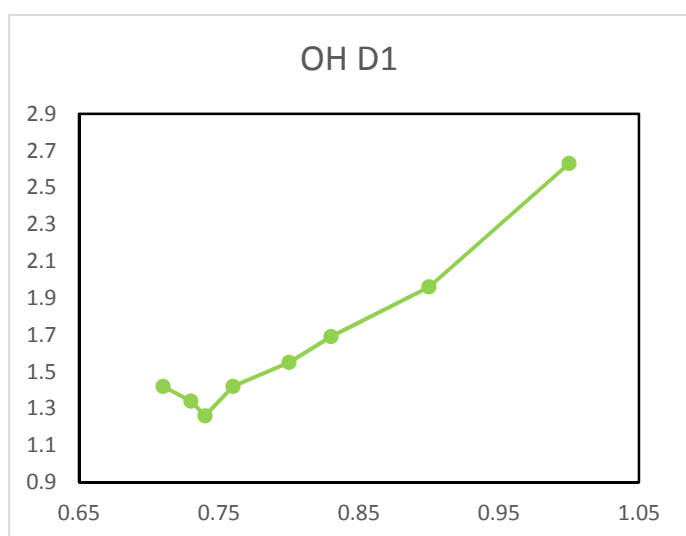
Gb/D3 1:12.27 $T_1: 1.42 \text{ sec} \pm 0.01281$ **D3** $T_1: 1.52 \text{ sec} \pm 0.007114$ 

3. Copies of T_1 exponential fitting by inversion recovery technique of other signals

3.1. T_1 exponential fitting of other signals

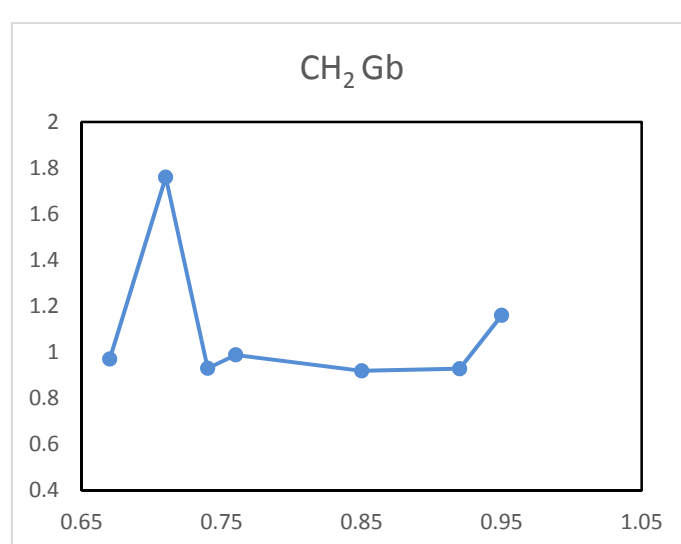
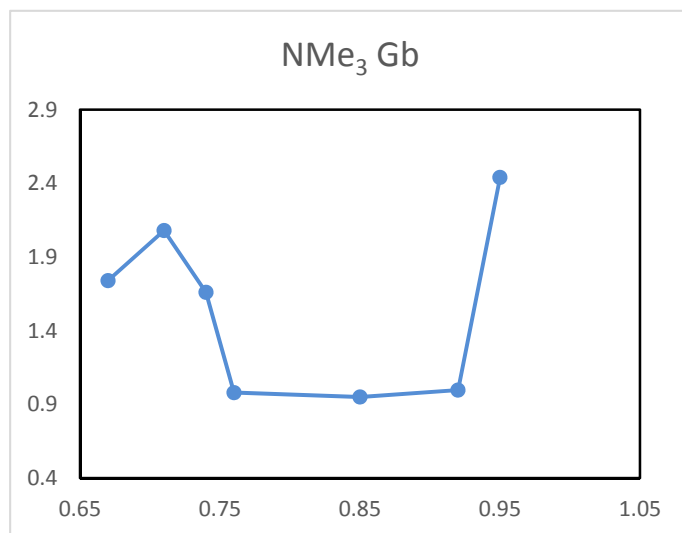
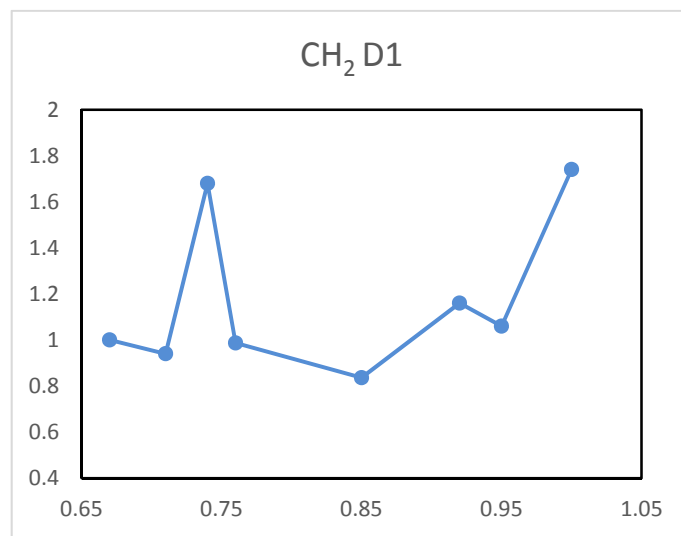
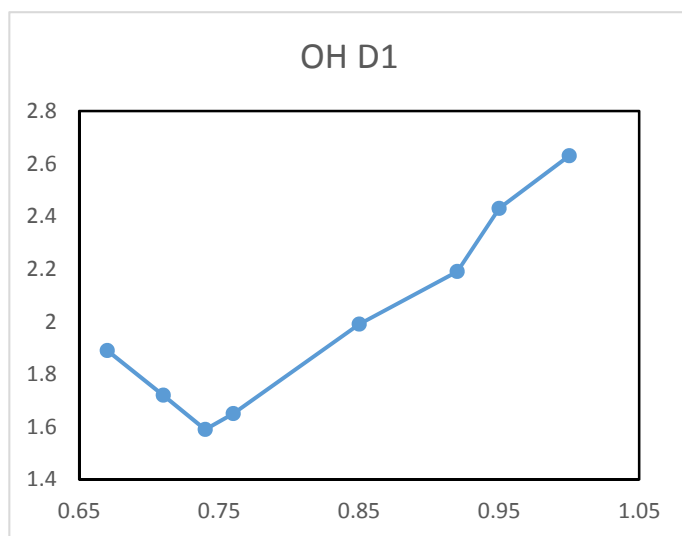
3.1. T_1 exponential fitting of Gb/D1 mixtures

Mixture	OH D1	CH ₂ D1	NMe ₃ Gb	CH ₂ Gb
Gb/D1 1:2.44	1.42	1.02	0.561	0.537
Gb/D1 1:2.73	1.34	1.04	0.58	0.555
Gb/D1 1:2.85	1.26	1.03	0.572	0.548
Gb/D1 1:3.18	1.42	0.583	0.61	0.585
Gb/D1 1:3.91	1.55	0.633	0.655	0.62
Gb/D1 1:4.85	1.69	0.84	0.718	0.701
Gb/D1 1:8.65	1.96	0.805	0.849	0.799
D1	2.63	1.74		



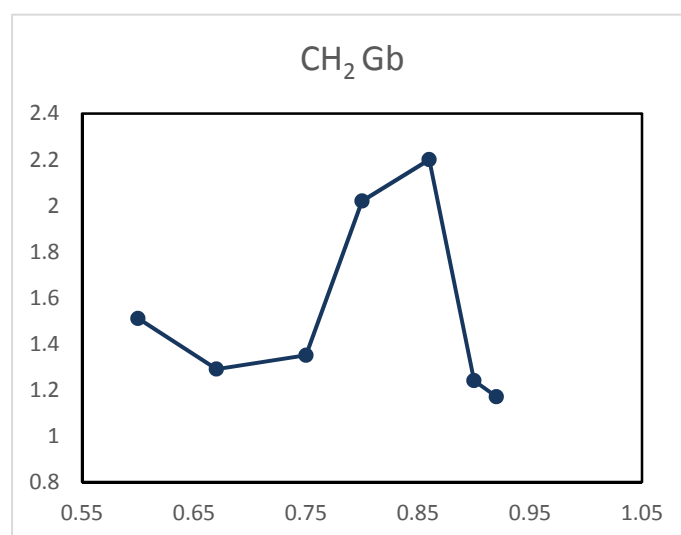
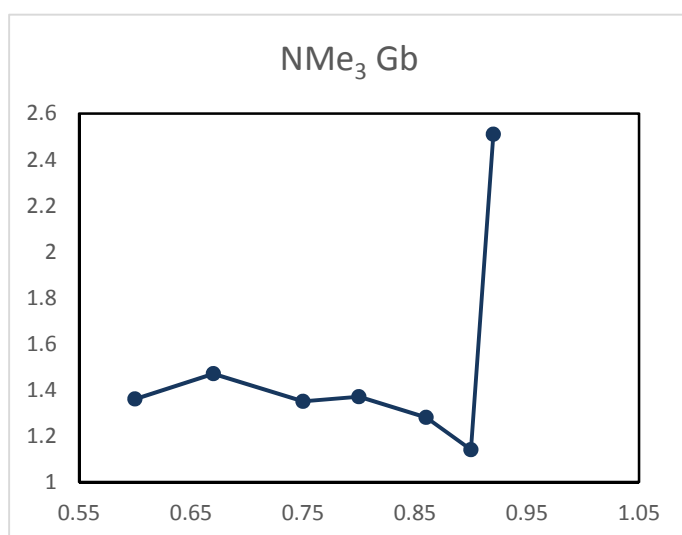
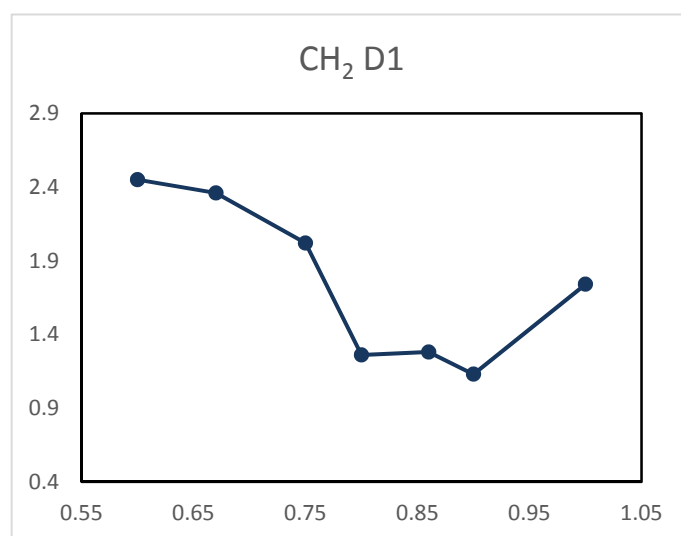
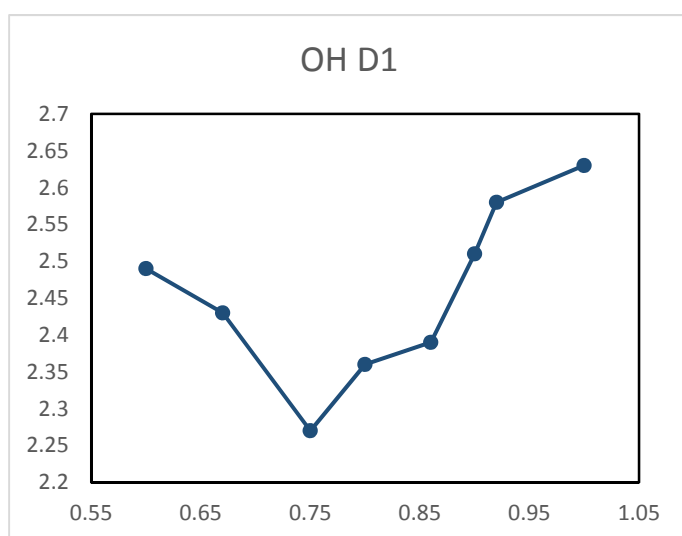
3.2. T_1 exponential fitting of Gb/D1 mixtures + 3 eq H₂O

Mixture	OH D1	CH ₂ D1	NMe ₃ Gb	CH ₂ Gb
Gb/D1 1:2.01	1.89	1	1.74	0.97
Gb/D1 1:2.49	1.72	0.94	2.08	1.76
Gb/D1 1:2.85	1.59	1.68	1.66	0.93
Gb/D1 1:3.11	1.65	0.987	0.98	0.988
Gb/D1 1:5.76	1.99	0.836	0.95	0.919
Gb/D1 1:11.81	2.19	1.16	0.997	0.928
Gb/D1 1:18.99	2.43	1.06	2.44	1.16
D1	2.63	1.74		



3.3. T_1 exponential fitting of Gb/D1 mixtures + 6 eq H₂O

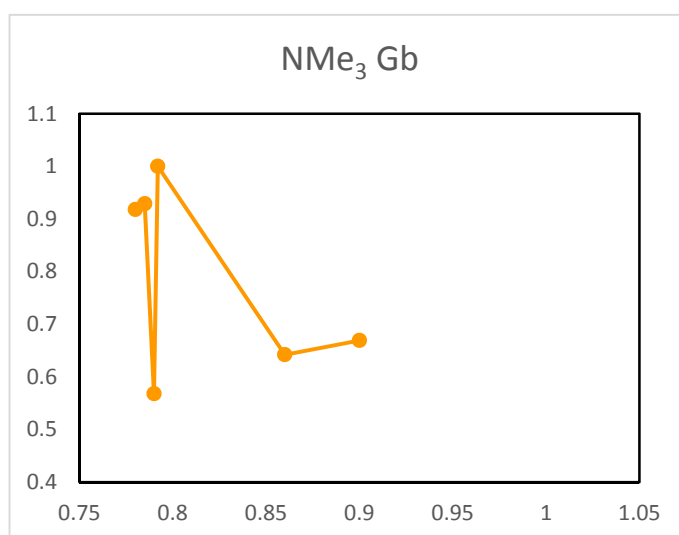
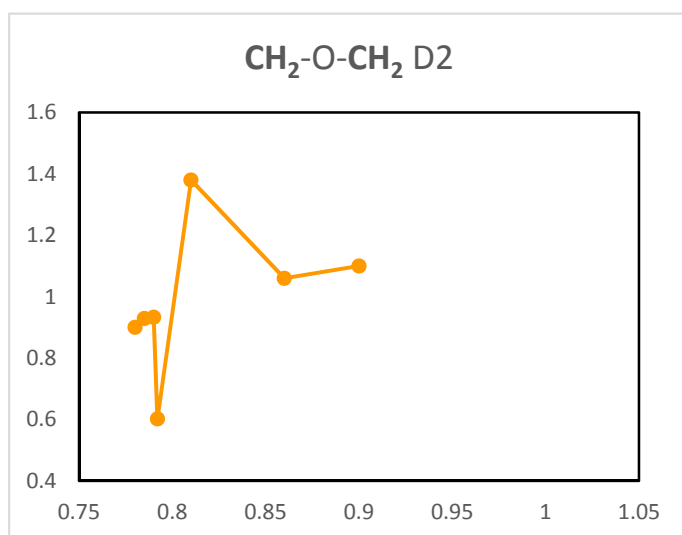
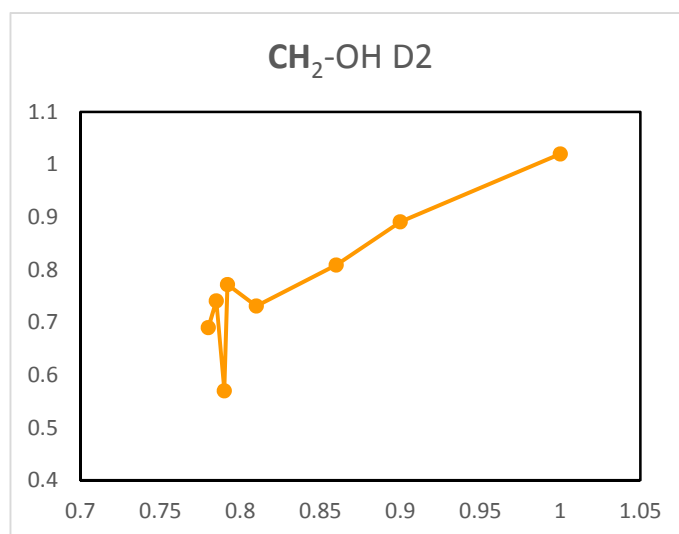
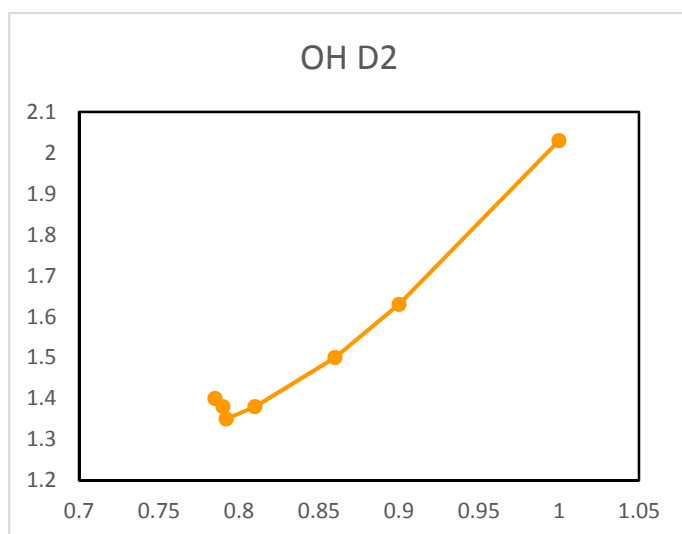
Mixture	OH D1	CH ₂ D1	NMe ₃ Gb	CH ₂ Gb
Gb/D1 1:1.52	2.49	2.45	1.36	1.51
Gb/D1 1:2.02	2.43	2.36	1.47	1.29
Gb/D1 1:3.02	2.27	2.02	1.35	1.35
Gb/D1 1:4.10	2.36	1.26	1.37	2.02
Gb/D1 1:6.05	2.39	1.28	1.28	2.2
Gb/D1 1:9.11	2.51	1.13	1.14	1.24
Gb/D1 1:11.82	2.58	*	2.51	1.17
D1	2.63	1.74		

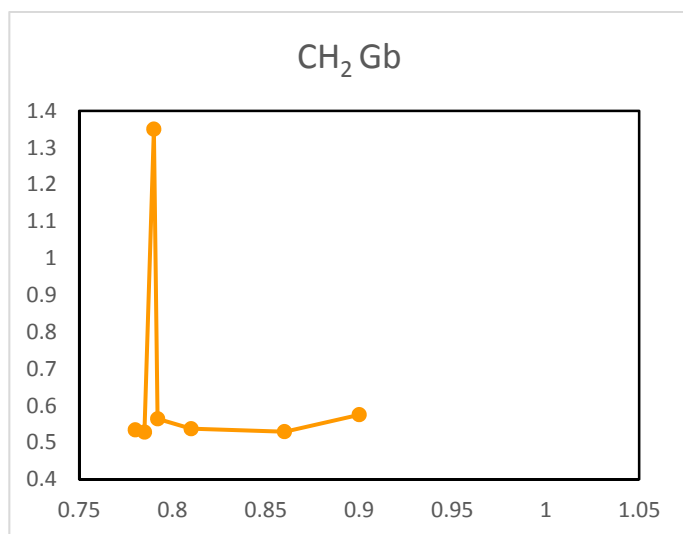


* error

3.4. T_1 exponential fitting of Gb/D2 mixtures

Mixture	OH D2	$\underline{\text{CH}_2\text{-O-CH}_2}$ D2	$\underline{\text{CH}_2\text{-OH}}$ D2	NMe ₃ Gb	CH ₂ Gb
Gb/D2 1:3.63	1.4	0.9	0.69	0.918	0.534
Gb/D2 1:3.66	1.38	0.929	0.741	0.929	0.528
Gb/D2 1:3.76	1.35	0.933	0.57	0.568	1.35
Gb/D2 1:3.80	1.53	0.602	0.772	1	0.564
Gb/D2 1:4.21	1.38	1.38	0.731	0.642	0.537
Gb/D2 1:6.07	1.5	1.06	0.809	0.669	1.13
Gb/D2 1:8.91	1.63	1.1	0.891	*	0.575
D2	2.03	*	1.02		

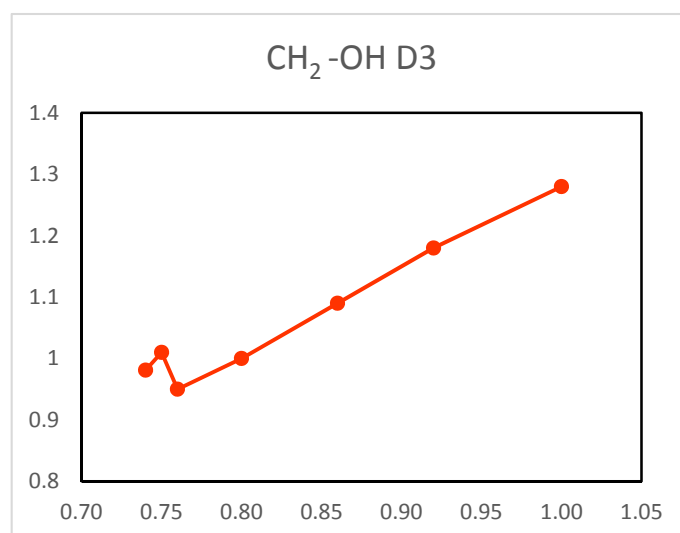
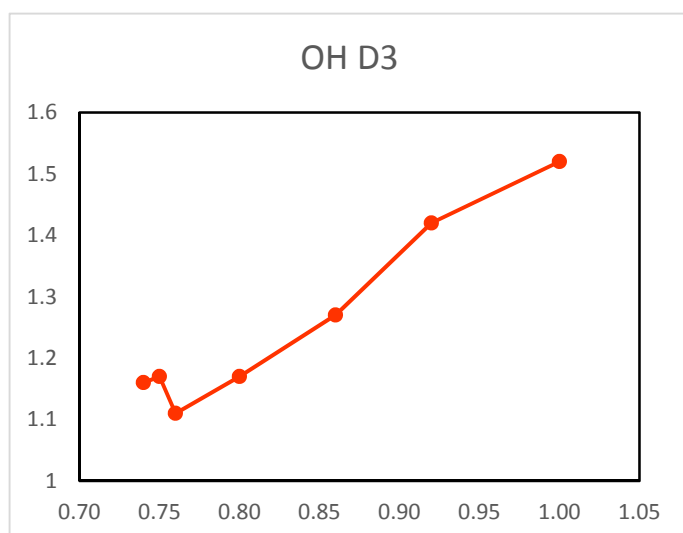


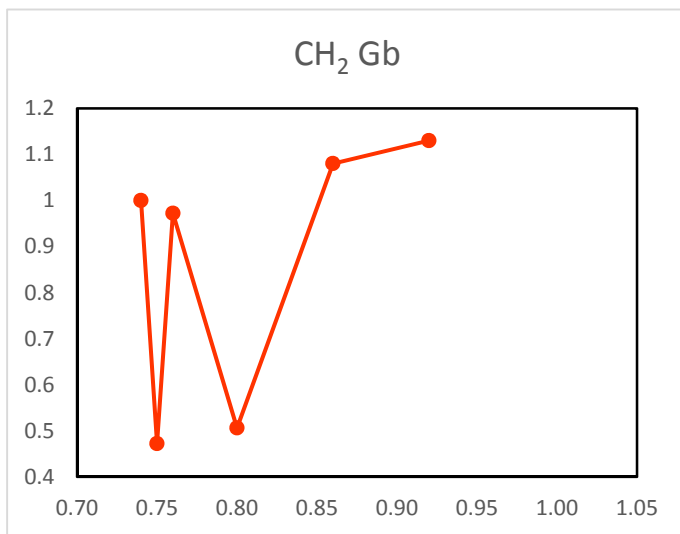
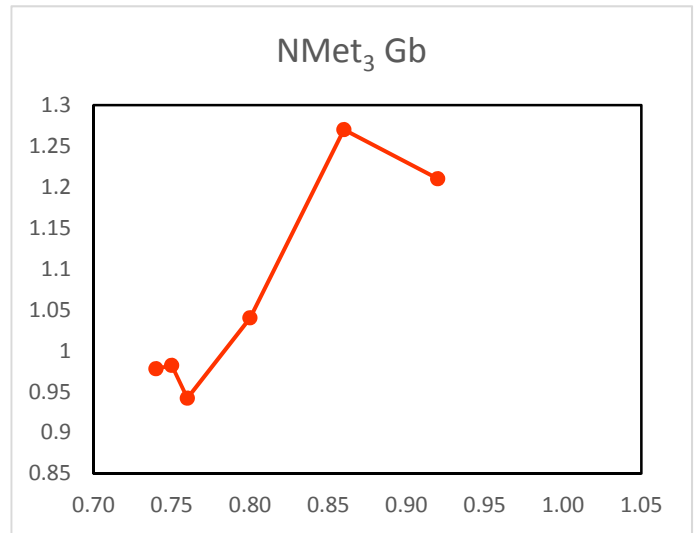
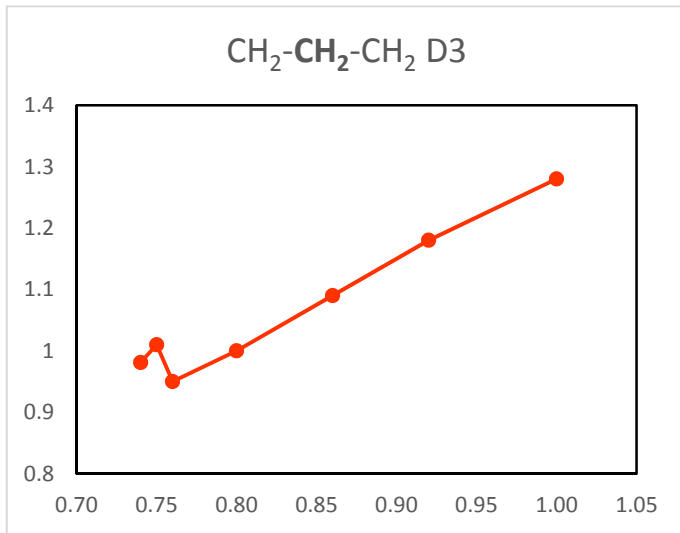


*error

3.5. T_1 exponential fitting of Gb/D3 mixtures

Mixture	OH D3	CH ₂ -OH D3	CH ₂ -CH ₂ -CH ₂ D3	NMe ₃ Gb	CH ₂ Gb
Gb/D3 1:2.86	1.16	0.988	0.981	0.978	1
Gb/D3 1:2.93	1.17	0.994	1.01	0.982	0.472
Gb/D3 1:3.10	1.11	0.95	0.95	0.942	0.972
Gb/D3 1:4.05	1.17	1	1	1.04	0.506
Gb/D3 1:6.32	1.27	1.09	1.09	1.27	1.08
Gb/D3 1:12.27	1.42	1.21	1.18	1.21	1.13
D3	1.52	1.27	1.28		

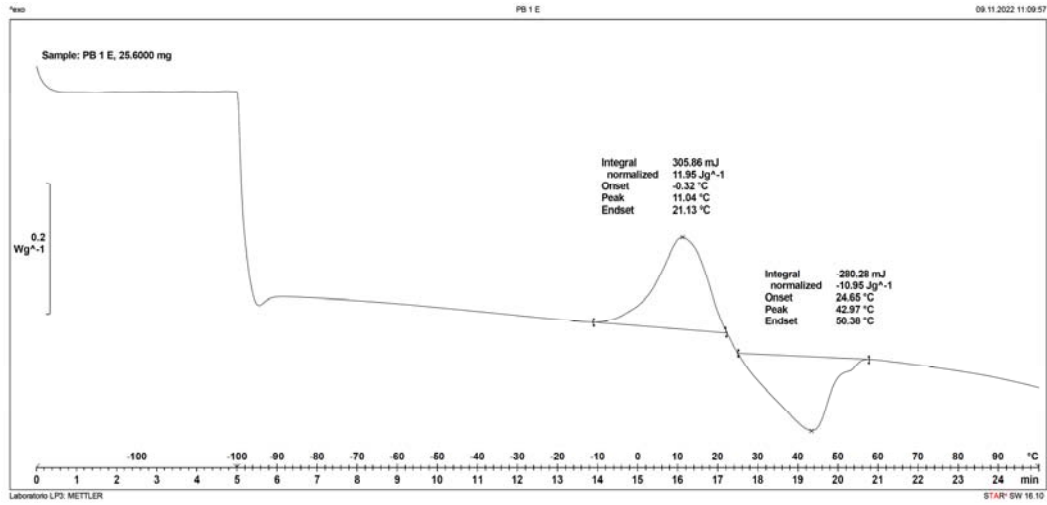




4. Copies of DSC plotting of Gb/D1 mixtures

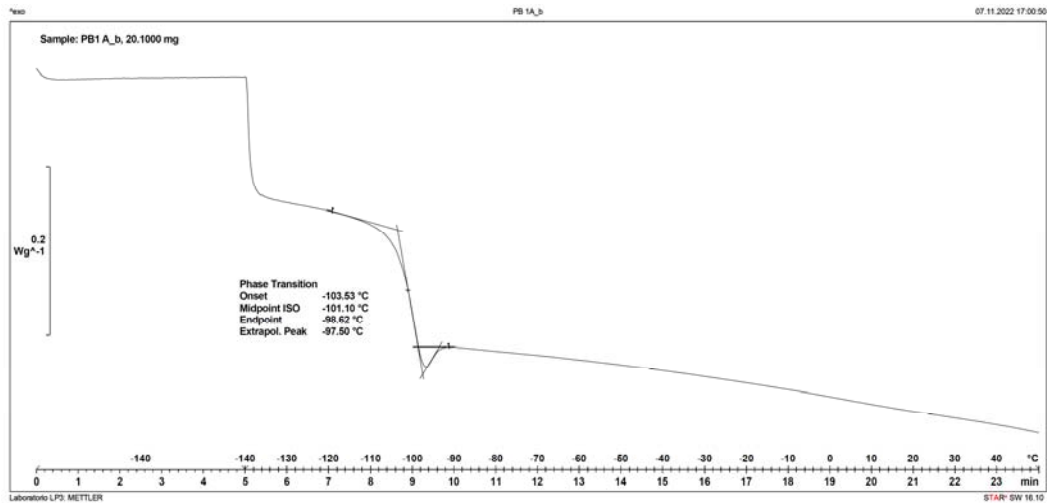
Gb/D1 1:2.23

XD= 0.69



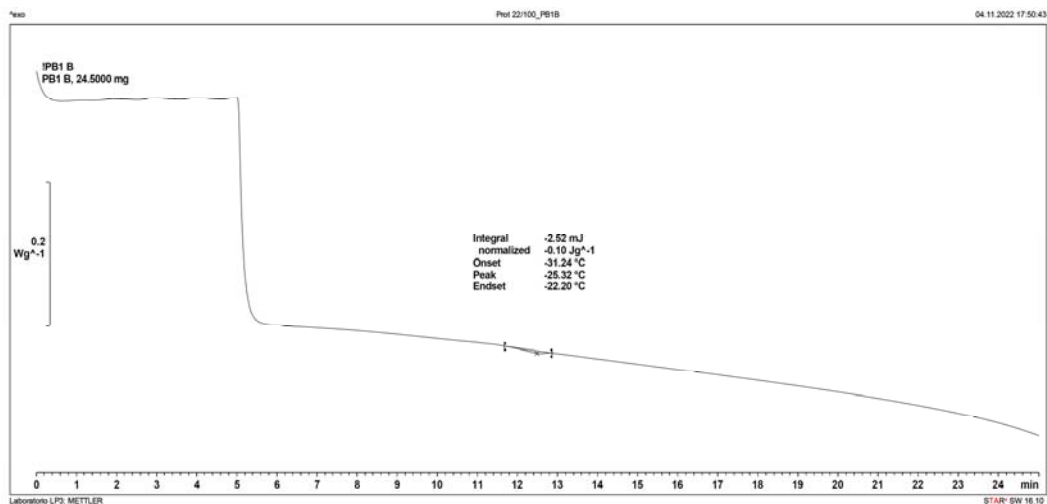
Gb/D1 1:2.85

XD= 0.74

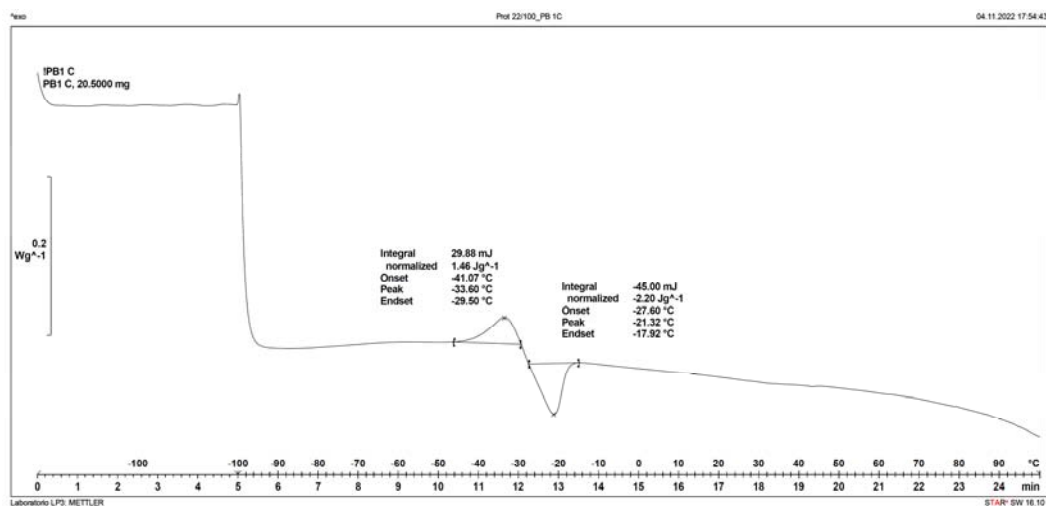


Gb/D1 1:4.97

XD= 0.83



Gb/D1 1:8
XD= 0.88



D1
XD= 1

