# Supplementary Materials

**Figure S1.** FT-IR spectrum of *N*-benzyl chitosan.

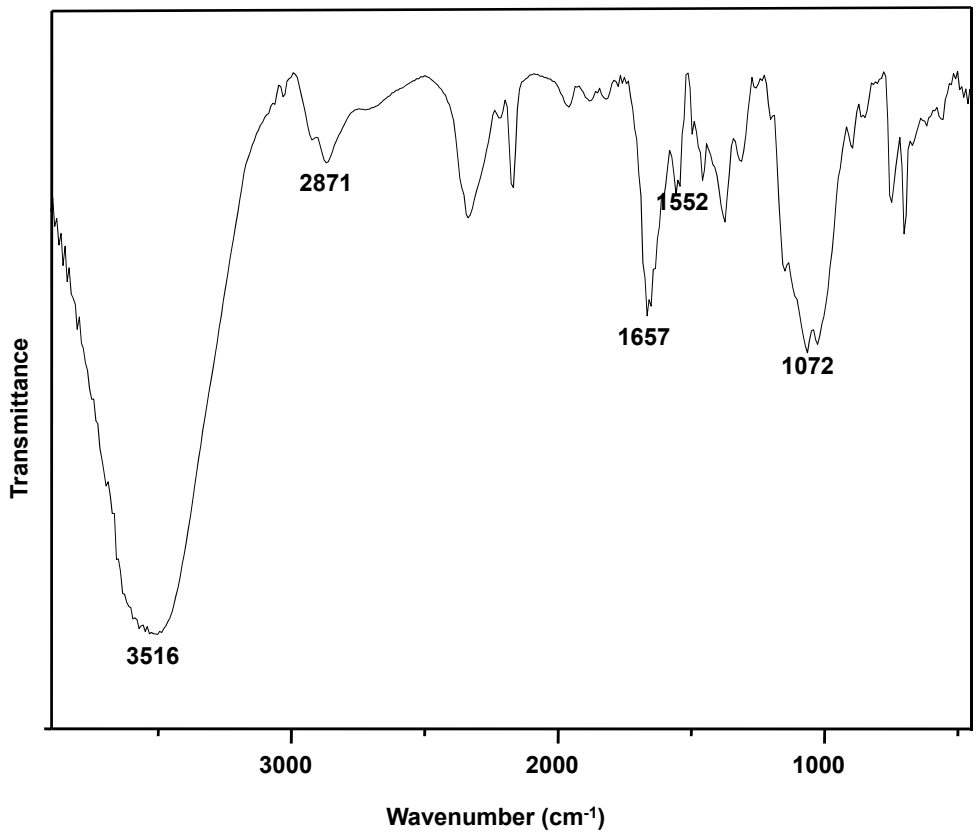


**Figure S2.** FT-IR spectrum of compound **1**.

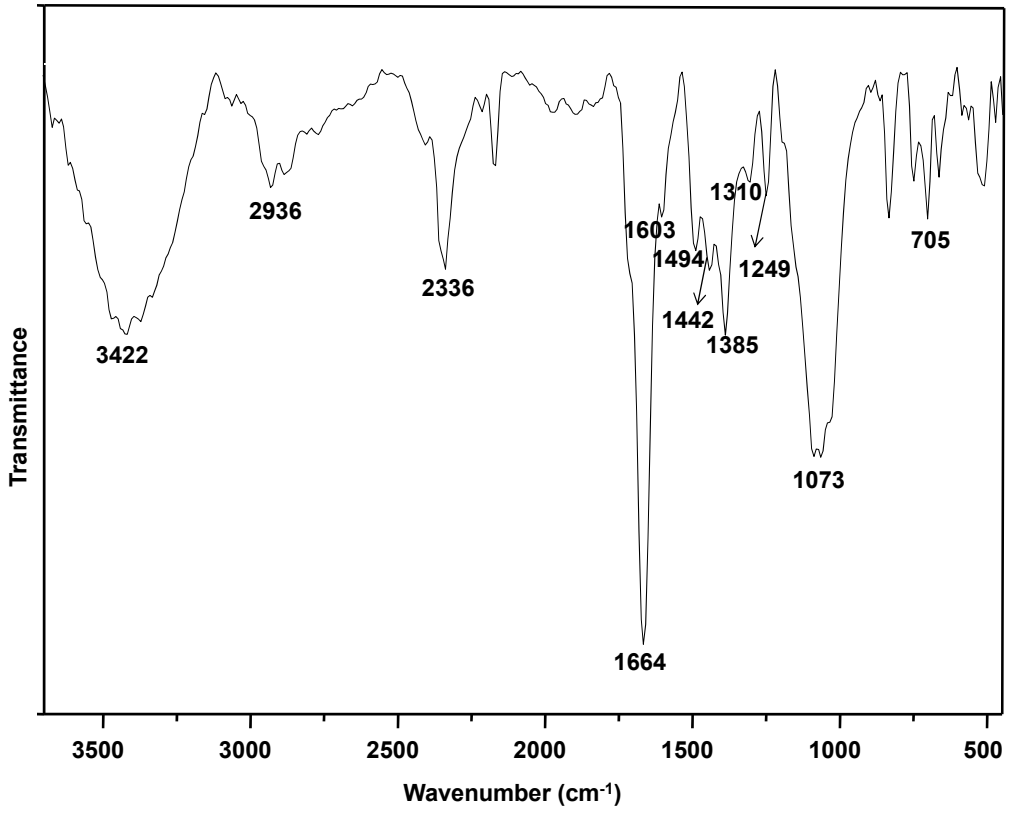

**Figure S3.** FT-IR spectrum of compound **2**.

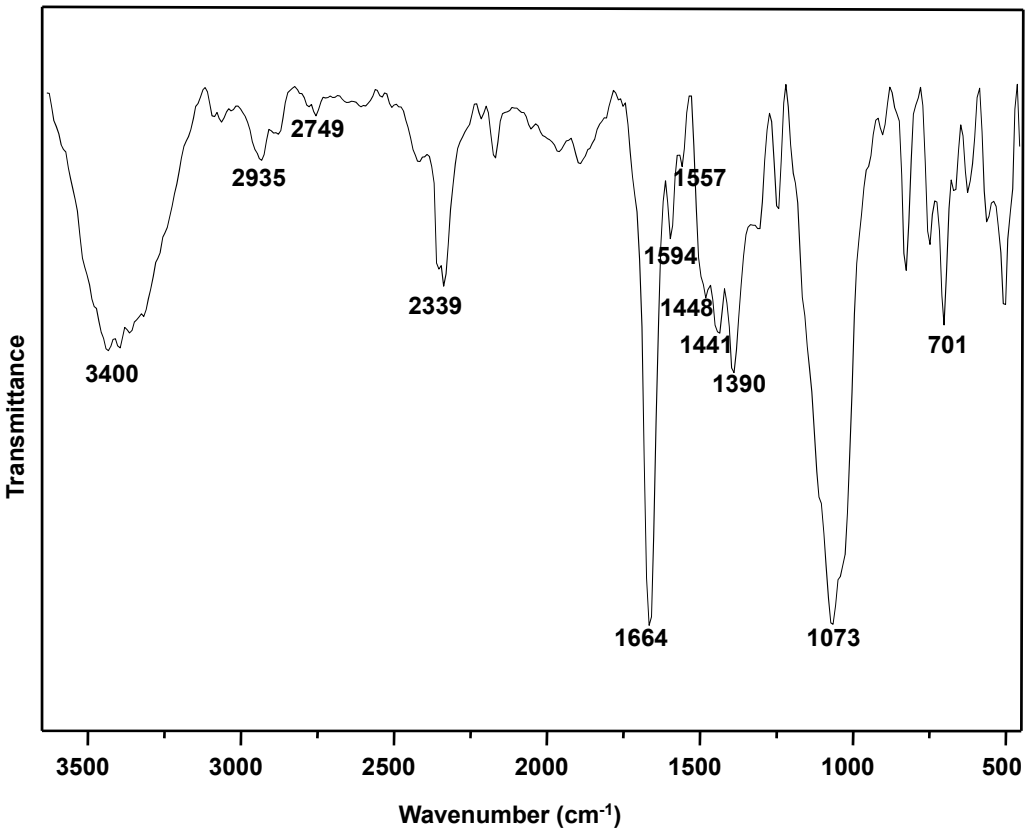


**Figure S4.** FT-IR spectrum of compound **3**.

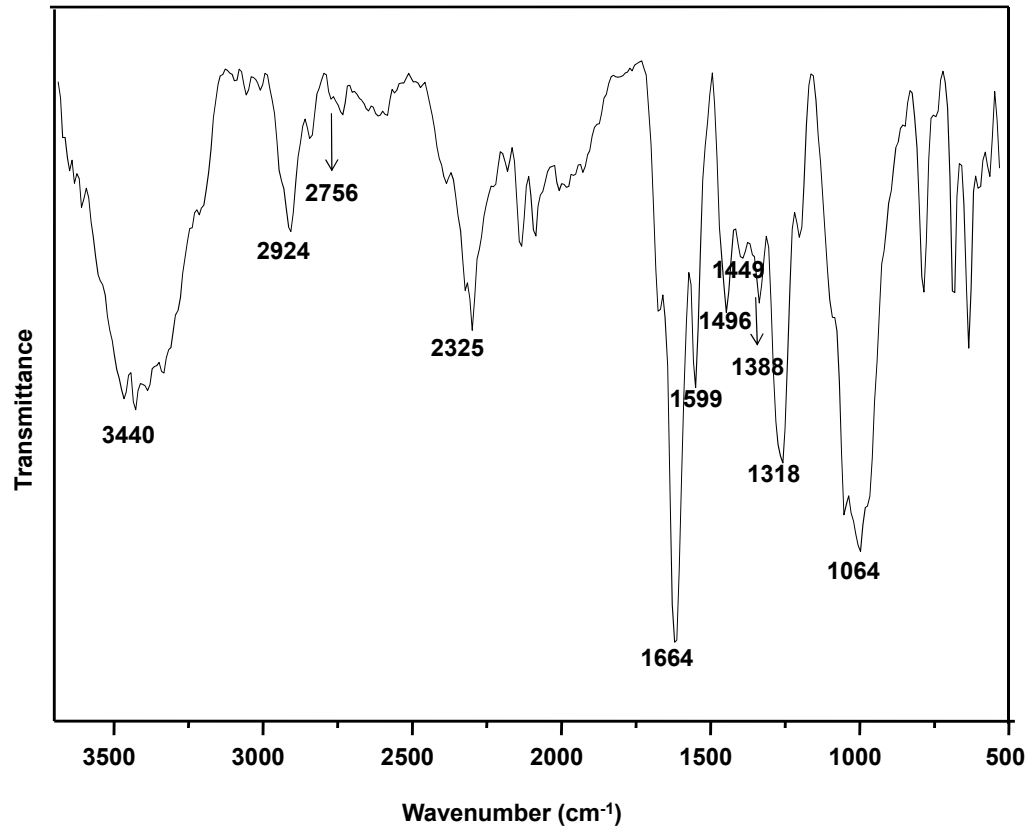

**Figure S5.** FT-IR spectrum of compound **4**.

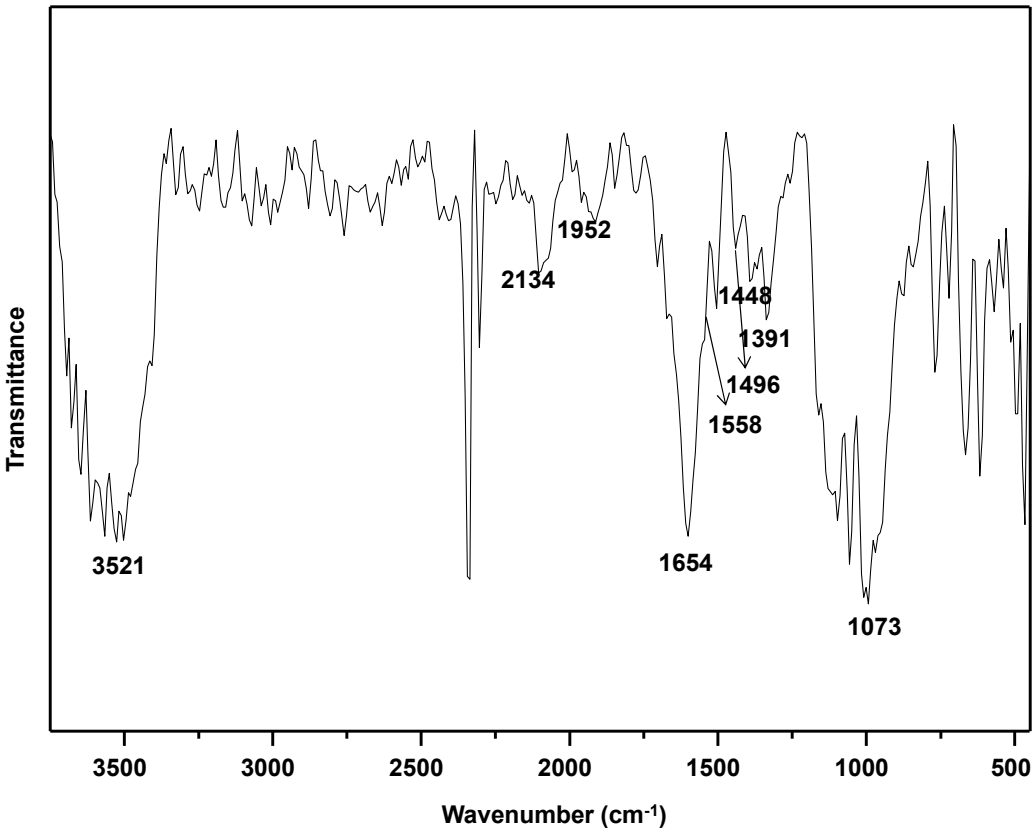


**Figure S6.** FT-IR spectrum of compound **5**.

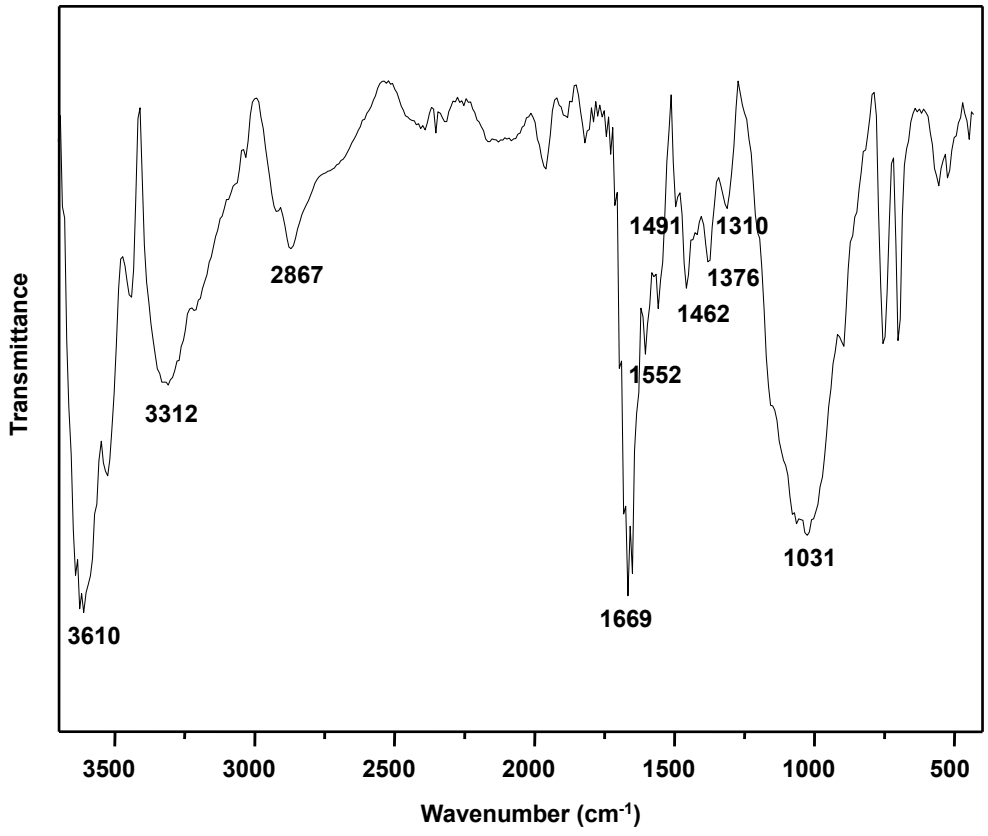

**Figure S7.** [1]H-NMR spectrum of chitosan.

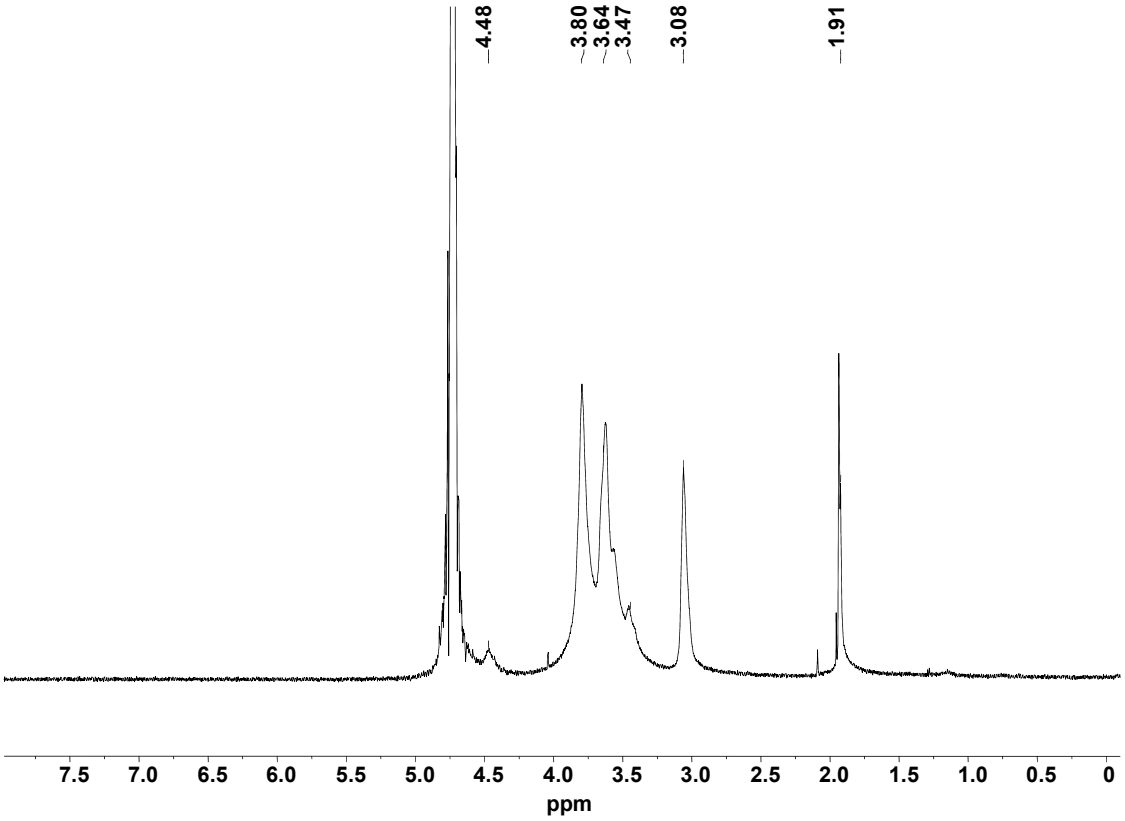


**Figure S8.** [1]H-NMR spectrum of *N*-benzyl chitosan.

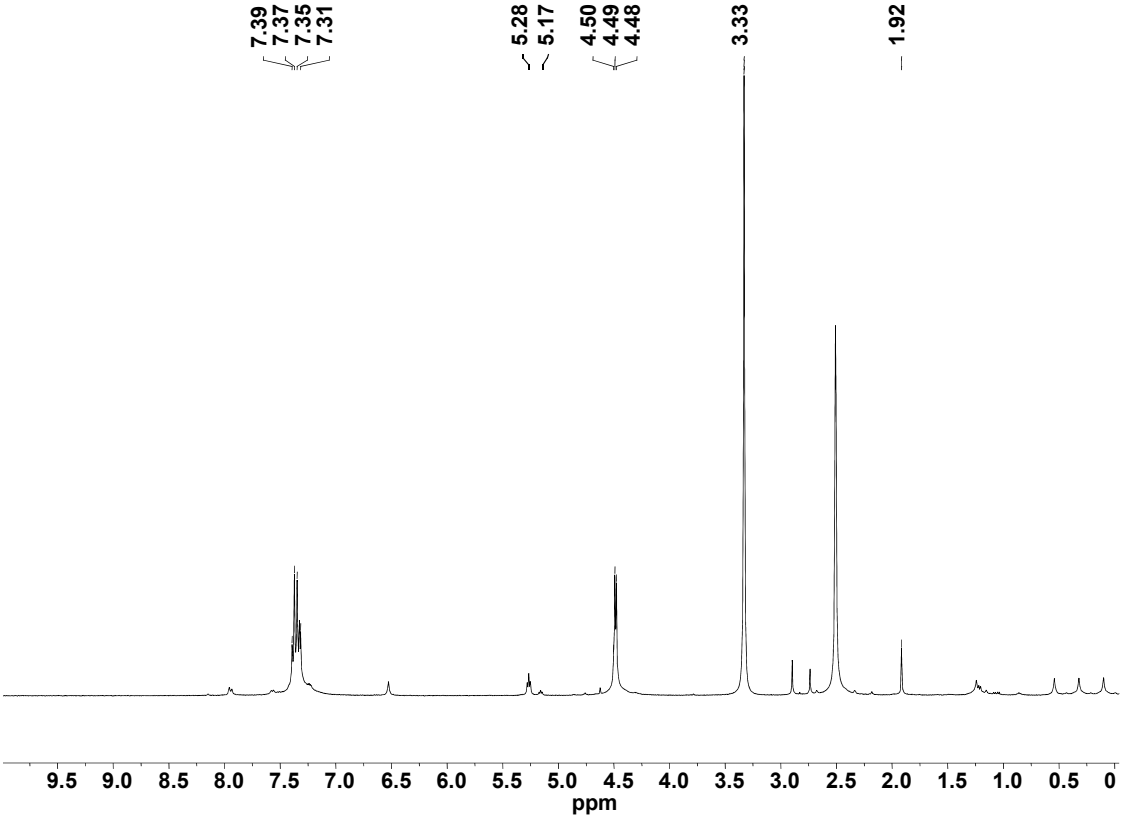

**Figure S9.** $^{1}$H-NMR spectrum of compound **1**.

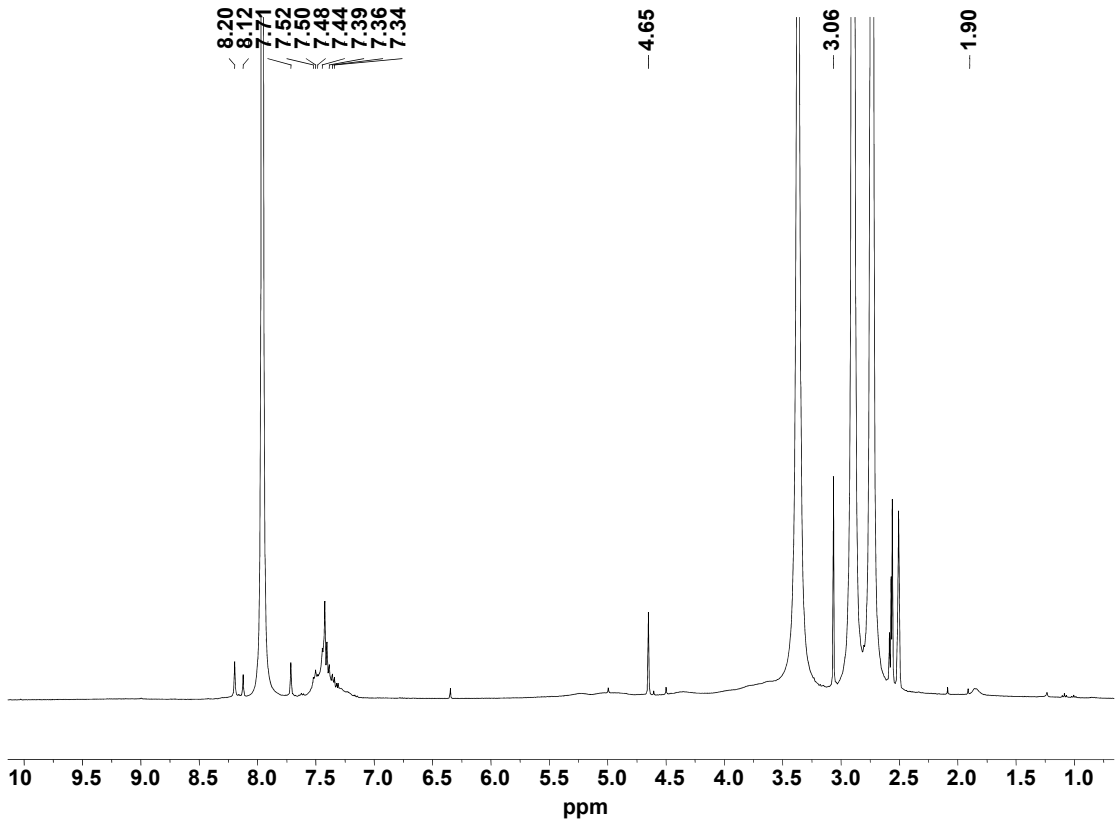


**Figure S10.** $^{1}$H-NMR spectrum of compound **2**.

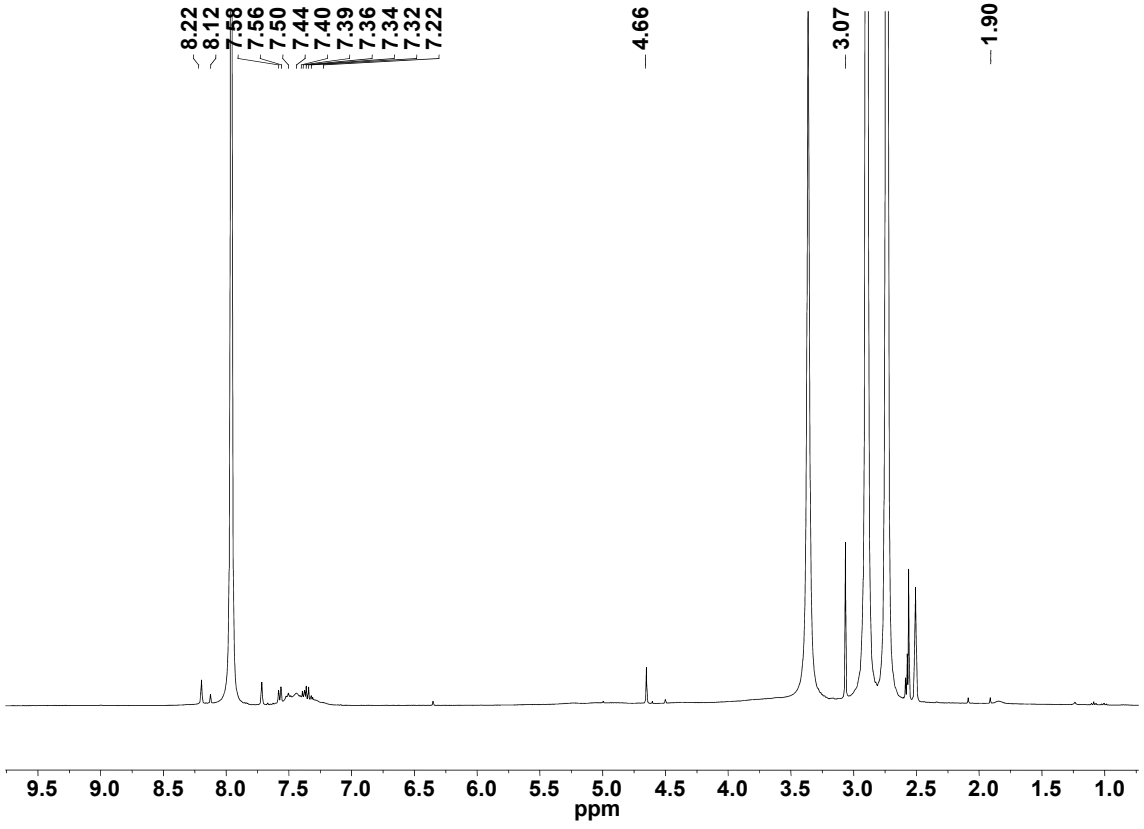

**Figure S11.** $^{1}$H-NMR spectrum of compound **3**.

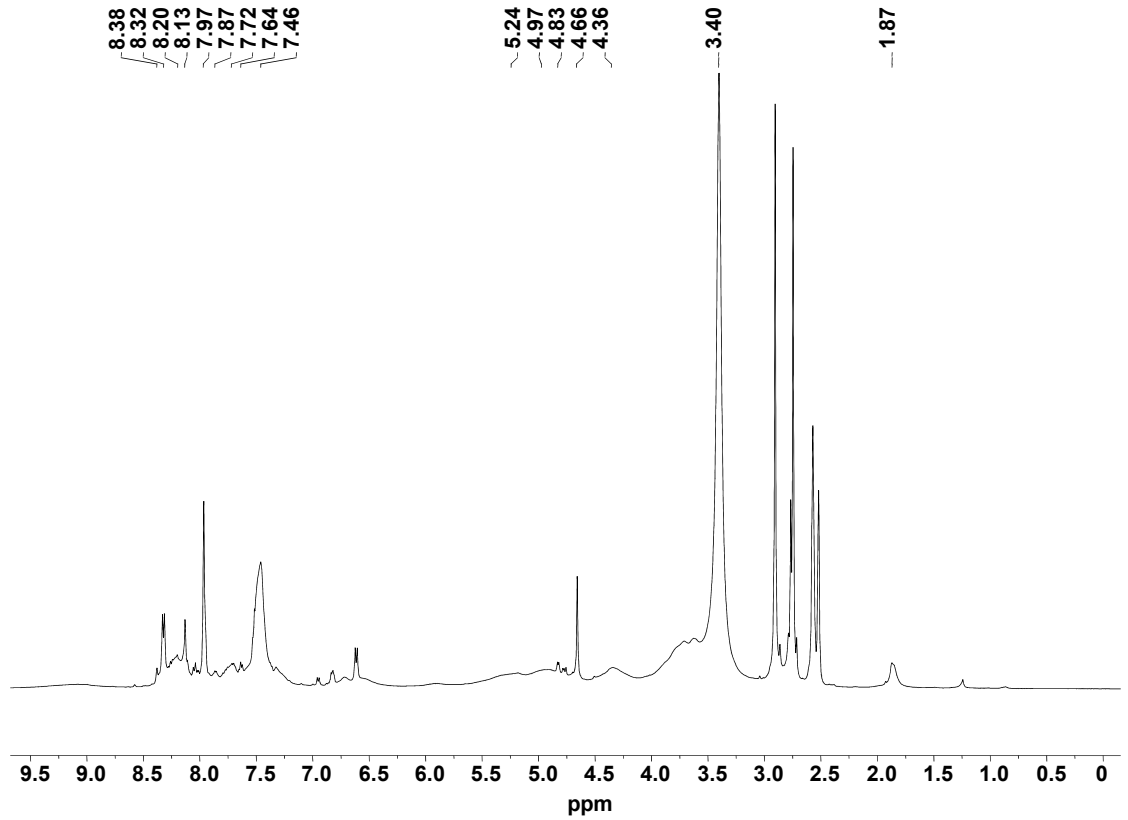


**Figure S12.** $^{1}$H-NMR spectrum of compound **4**.

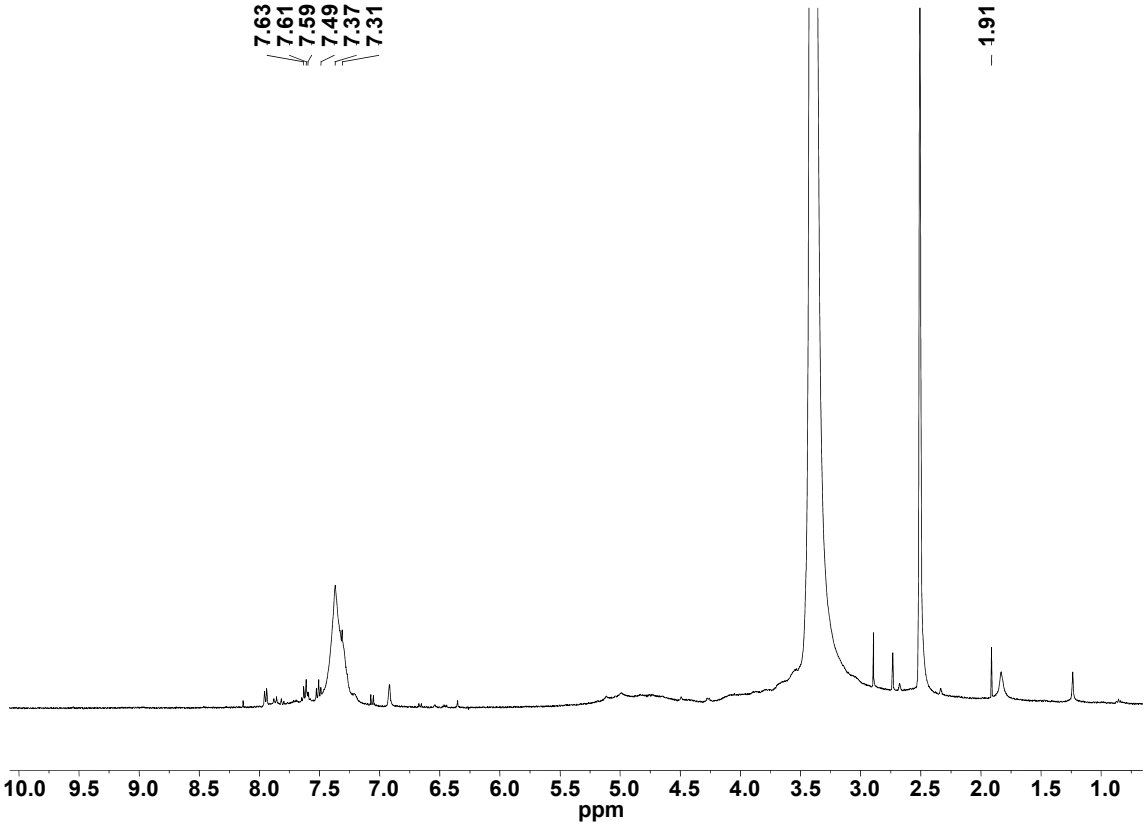

**Figure S13.** $^{1}$H-NMR spectrum of compound **5**.

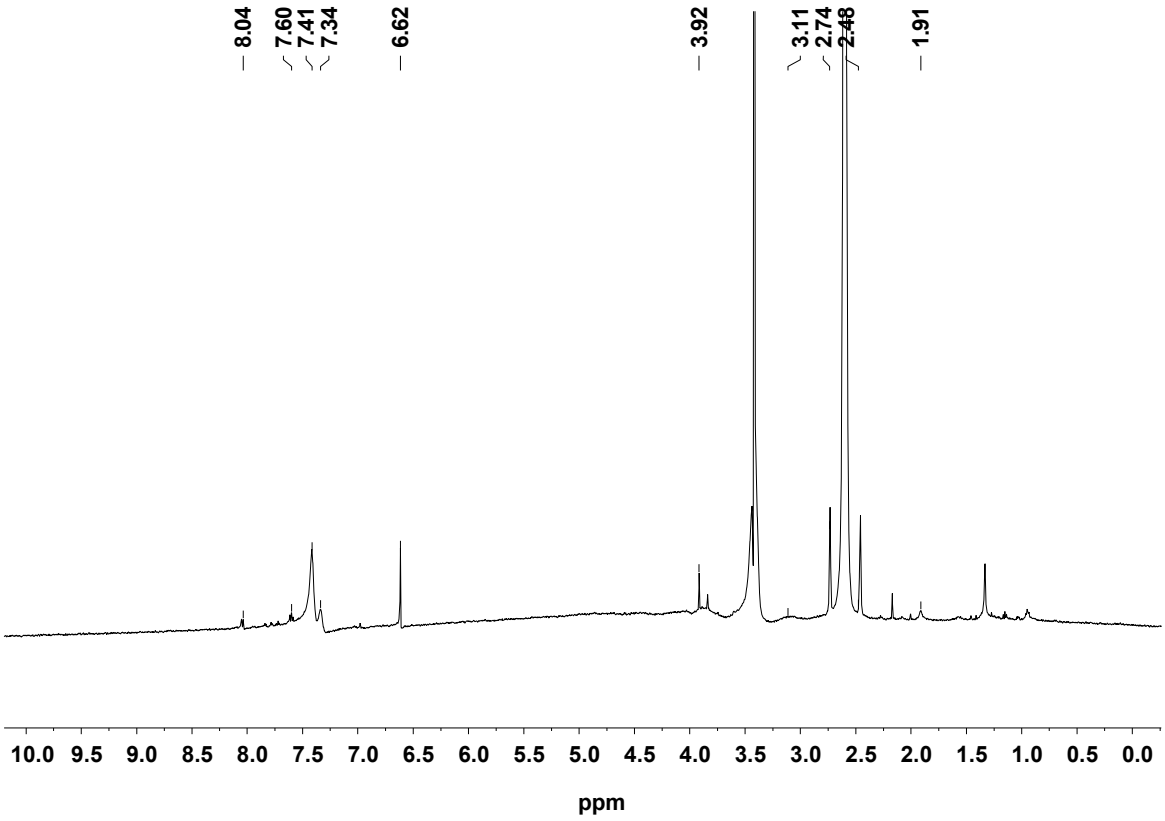


**Figure S14.** $^{15}$N-NMR spectrum of chitosan.

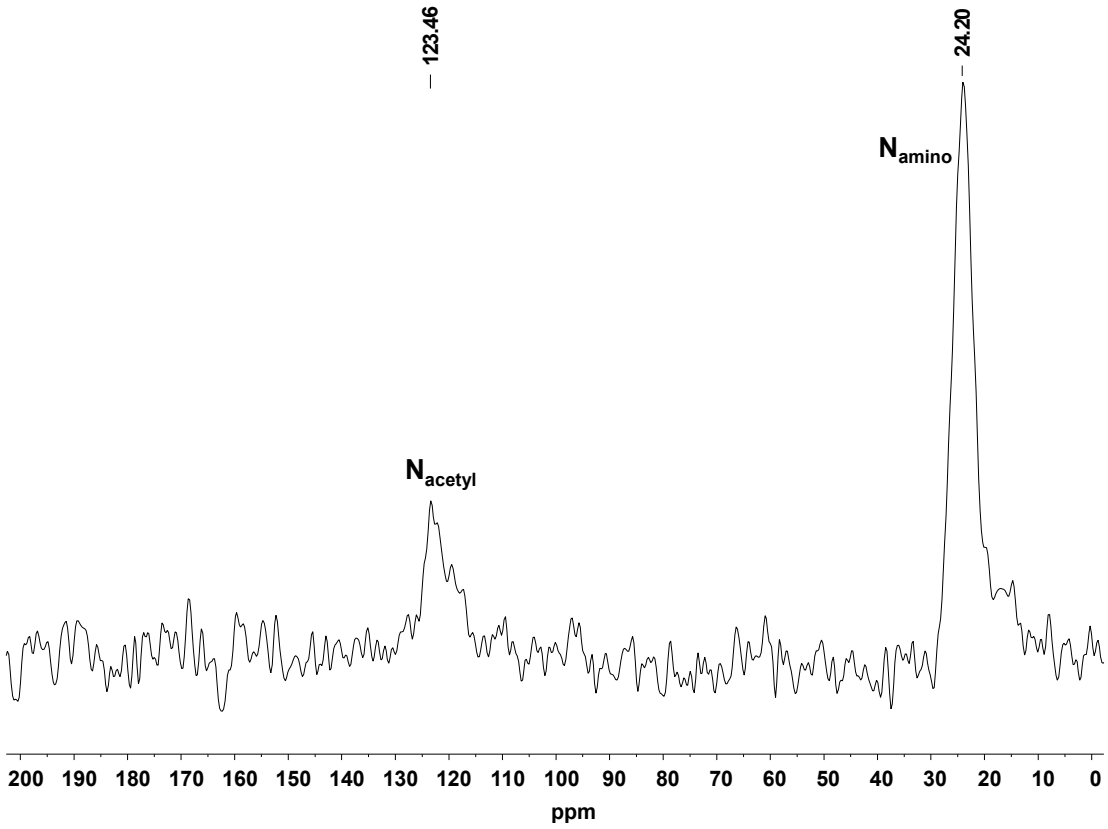

**Figure S15.** $^{15}$N-NMR spectrum of *N*-benzyl chitosan.

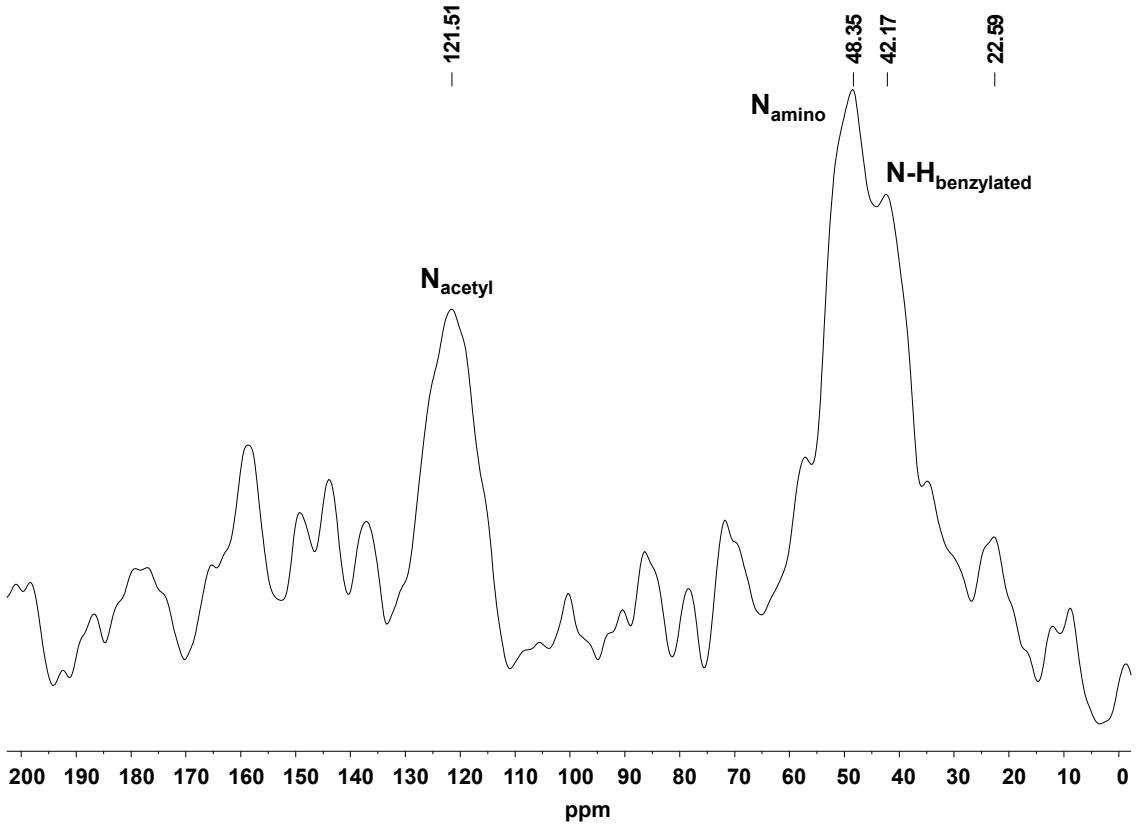


**Figure S16.** $^{15}$N-NMR spectrum of compound **5**.

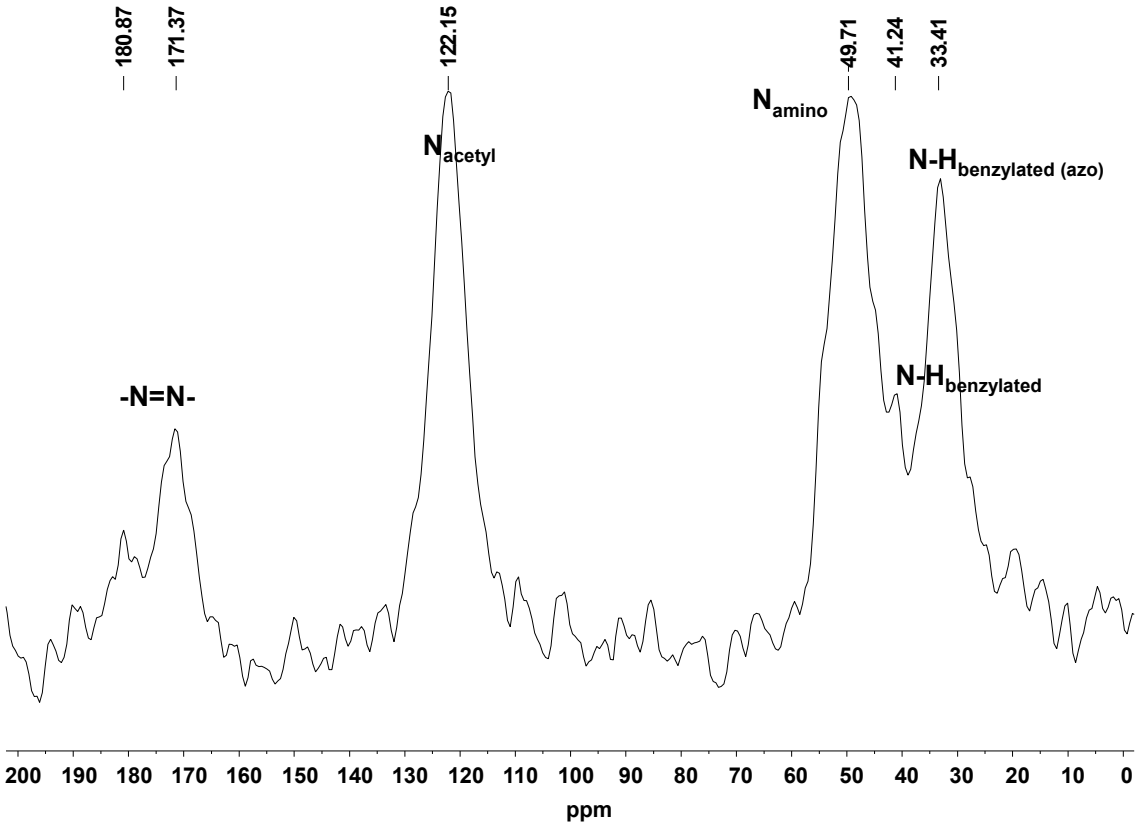

**Table S1.** Calculated areas for determination of the degree of deacetylation (DD) of chitosan. The $^{15}$N-NMR measurements were performed in triplicate.

| Signal (ppm) | Chitosan | | | Average | Error |
|---|---|---|---|---|---|
| 24.20 | 2,644,436 | 2,644,446 | 2,644,456 | 2,644,446 | 10 |
| 123.46 | 707,978 | 707,988 | 707,998 | 707,988 | 10 |

**Table S2.** Calculated areas for determination of the degree of substitution (DS) of *N*-benzyl chitosan. The $^{15}$N-NMR measurements were performed in triplicate.

| Signal (ppm) | *N*-benzyl Chitosan | | | Average | Error |
|---|---|---|---|---|---|
| 42.17 | 854,556 | 854,666 | 854,676 | 854,666 | 66 |
| 48.35 | 1,512,335 | 1,512,325 | 1,512,345 | 1,512,325 | 10 |
| 123.46 | 1,158,029 | 1,158,039 | 1,158,049 | 1,158,039 | 10 |

**Table S3.** Calculated areas for determination of the degree of substitution (DS) of compound **5**. The $^{15}$N-NMR measurements were performed in triplicate.

| Signal (ppm) | Compound 5 | | | Average | Error |
|---|---|---|---|---|---|
| 33.41 | 3,308,711 | 3,308,701 | 3,308,722 | 3,308,701 | 10 |
| 42.17 | 1,045,387 | 1,045,397 | 1,045,498 | 1,045,397 | 61 |
| 48.35 | 6,553,375 | 6,553,475 | 6,553,585 | 6,553,485 | 105 |
| 123.46 | 4,430,778 | 4,430,788 | 4,430,798 | 4,430,788 | 10 |
| 171.37 | 1,550,330 | 1,550,340 | 1,550,350 | 1,550,340 | 10 |
| 180.87 | 277,482 | 277,502 | 277,522 | 277,502 | 20 |

The degree of deacetylation (DD) and substitution (DS) can be calculated by $^{15}$N-NMR as follows:

$$DD = A_{NH2}/(A_{NH2} + A_{N\text{-}acteyl}) \times 100\% \quad (1)$$

$A_{NH2}$ and $A_{N\text{-}acteyl}$ correspond to the integral areas of amino and acetyl groups, respectively.

The DD and DS were calculated using the average value of the areas of each $^{15}$N-NMR signal.

Using Equation (1), the DD of chitosan was 78%. The DS of *N*-benzyl chitosan was 56% and compound **5** was 42%.

The degree of deacetylation for chitosan by $^{1}$H-NMR technique was determined using the equation:

$$DD\ (\%) = 1 - (1/3\ I_{CH3}/I_{(H2\text{-}GlyN)}) \times 100$$

where $I_{CH3}$ is integral of -$CH_3$ signal and $I_{(H2\text{-}GlyN)}$ is the integral of the proton of C-2 carbon of GlyN.

The DD value obtained was 78%, which is in agreement with the DD value obtained by $^{15}$N-NMR technique.

The degree of substitution of the derivatives was also calculated by $^{1}$H-NMR technique using the equation:

$$DS\ (\%) = (Ar/n/I_{H2+H2'} + 1/3\ I_{CH3}) \times 100$$

where Ar is the integral of aromatic protons, n is number of hydrogen atom per substituent, $I_{H2+H2'}$ are integrals of the proton of C-2 carbon of GlyN, and $I_{CH3}$ is the integral of GlyNAc proton.

The obtained DS values are listed in the table below.

**Table S4.** Degree of substitution (DS) of *N*-benzyl chitosan and compounds **1**–**5**.

| Compounds | DS (%) |
|---|---|
| *N*-benzyl chitosan | 50 |
| 1 | 66 |
| 2 | 31 |
| 3 | 51 |
| 4 | 52 |
| 5 | 46 |

The integral areas of the proton signals are given in the Tables below.

**Table S5.** Calculated integrals average for determination of the degree of substitution (DS) of chitosan. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | Chitosan Integrals Average | Error |
|---|---|---|
| 1.93 | 2.43 | 10 |
| 3.06 | 3.89 | 10 |

**Table S6.** Calculated integrals average for determination of the degree of substitution (DS) of *N*-benzyl chitosan. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | *N*-benzyl Chitosan Integrals Average | Error |
|---|---|---|
| 2.00 | 1.62 | 50 |
| 3.13–3.19 | 2.97 | 22 |
| 7.36–7.89 | 6.18 | 60 |

**Table S7.** Calculated integrals average for determination of the degree of substitution (DS) of compound **1**. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | Compound 1 Integrals Average | Error |
|---|---|---|
| 1.84 | 3.10 | 60 |
| 3.54–3.68 | 13.88 | 10 |
| 7.36–7.89 | 21.53 | 10 |

**Table S8.** Calculated integrals average for determination of the degree of substitution (DS) of compound **2**. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | Compound 2 Integrals Average | Error |
|---|---|---|
| 1.85 | 1.16 | 50 |
| 3.71–3.76 | 3.68 | 22 |
| 7.12–8.24 | 21.53 | 60 |

**Table S9.** Calculated integrals average for determination of the degree of substitution (DS) of compound **3**. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | Compound 3 Integrals Average | Error |
|---|---|---|
| 1.87 | 1.03 | 38 |
| 3.61–3.67 | 4.11 | 42 |
| 7.32–8.38 | 18.40 | 10 |

**Table S10.** Calculated integrals average for determination of the degree of substitution (DS) of compound **4**. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | Compound 4 Integrals Average | Error |
|---|---|---|
| 1.83 | 5.35 | 10 |
| 3.053–3.16 | 10.41 | 10 |
| 7.25–8.24 | 54.50 | 10 |

**Table S11.** Calculated integrals average for determination of the degree of substitution (DS) of compound **5**. The $^{1}$H-NMR measurements were performed in triplicate.

| Signal (ppm) | Compound 5 Integrals Average | Error |
|---|---|---|
| 1.92 | 1.30 | 40 |
| 3.83–3.92 | 4.64 | 34 |
| 7.33–7.60 | 15.64 | 67 |

**Figure S17.** UV-Vis spectra of compounds **1**–**5**.

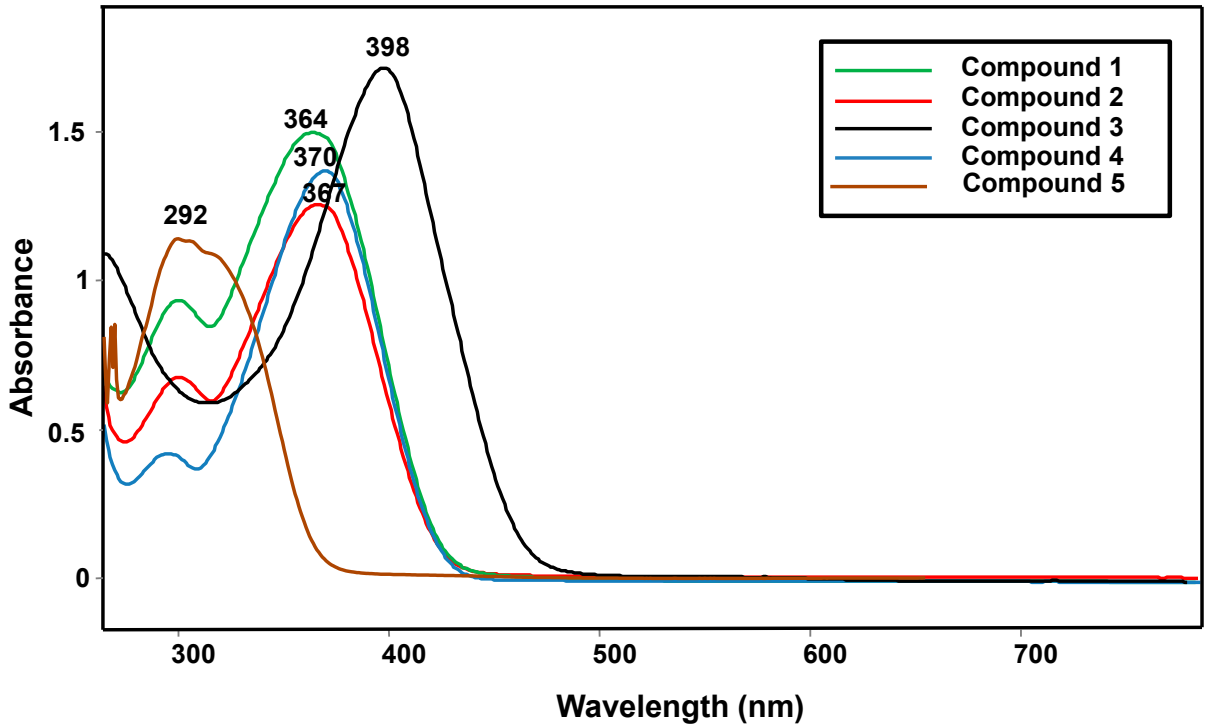

**Figure S18.** ESI(+)MS spectrum of chitosan hydrolysates.

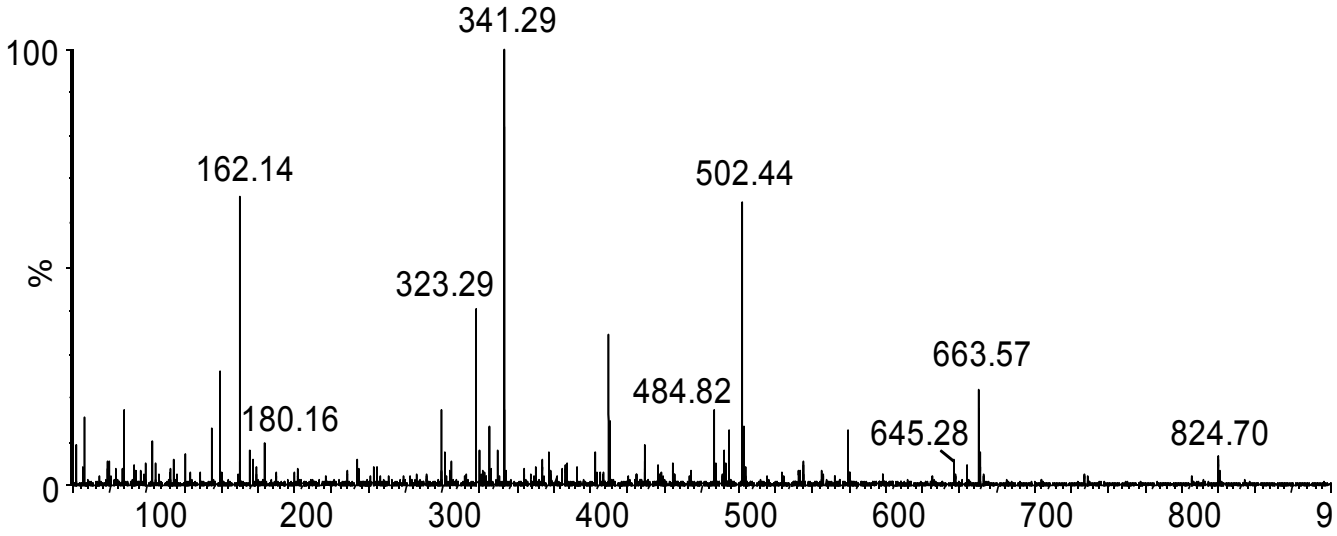


**Figure S19.** ESI(+)MS spectrum from the hydrolysis reaction mixture of compound **4** (10 M HCl).

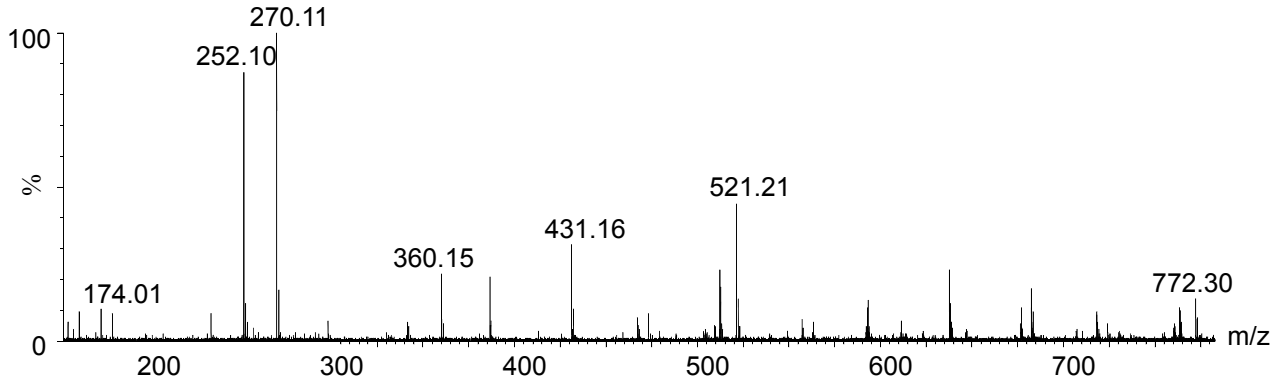


**Figure S20.** ESI(+)MS spectrum from the hydrolysis reaction mixture of compound **5** (10 M HCl).

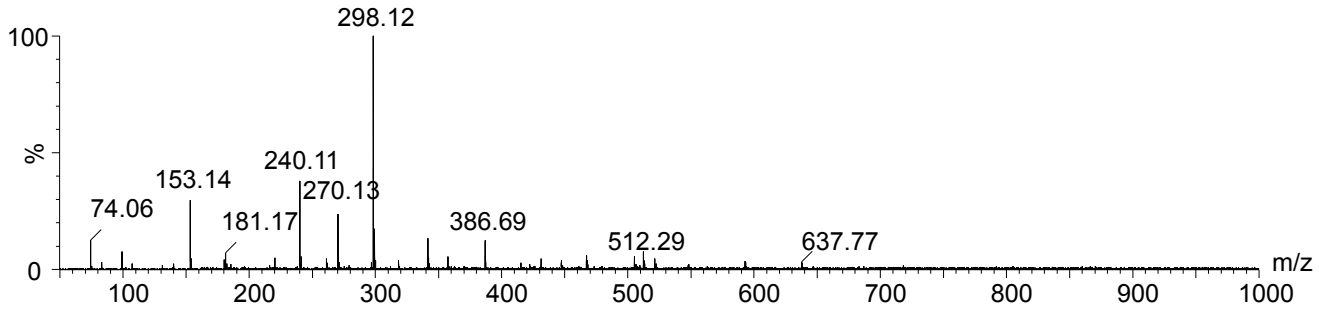

**Figure S21.** GC-MS spectrum from the hydrolysis reaction mixture of *p*-bromoaniline (10 M HCl).

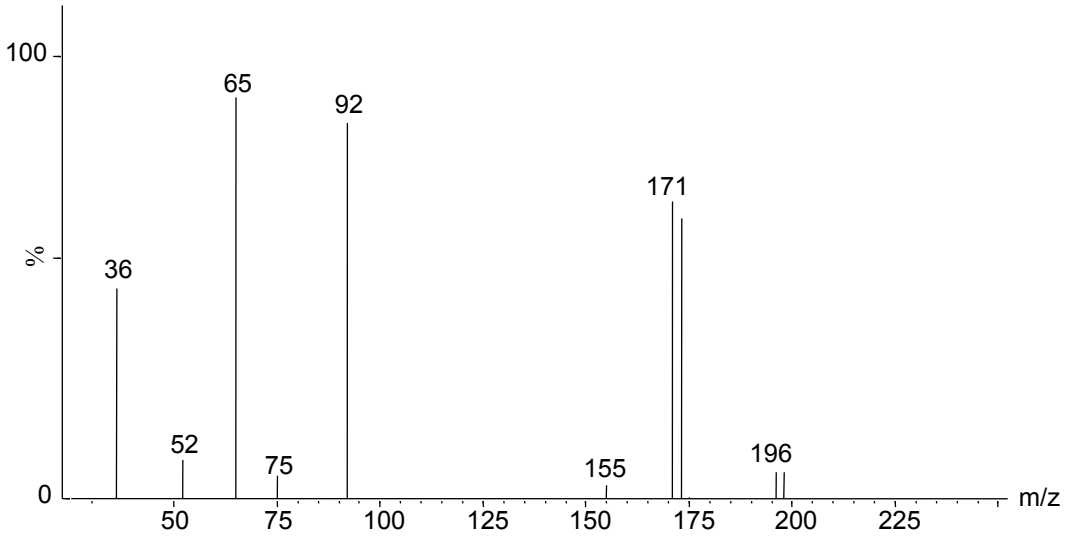


**Figure S22.** GC-MS spectrum from the hydrolysis reaction mixture of *p*-bromobenzenediazonium tetrafluorborate (10 M HCl).