

Figure S1.  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) spectrum of compound 1.

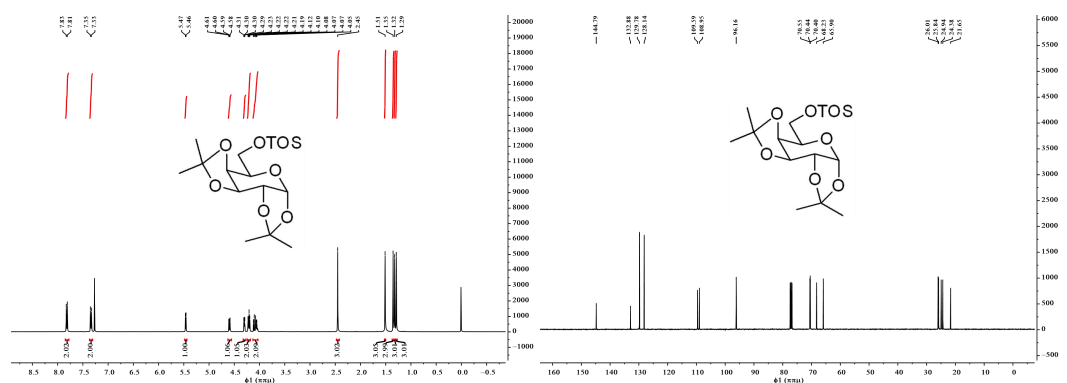


Figure S2.  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) spectrum of compound 2.

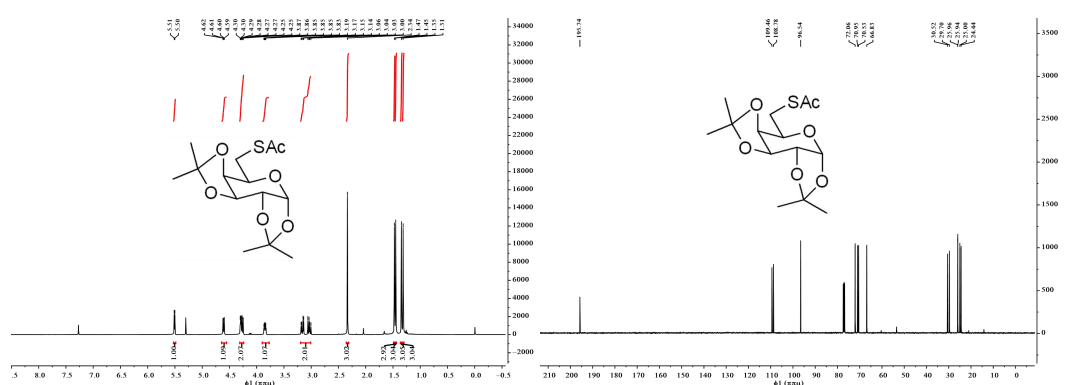
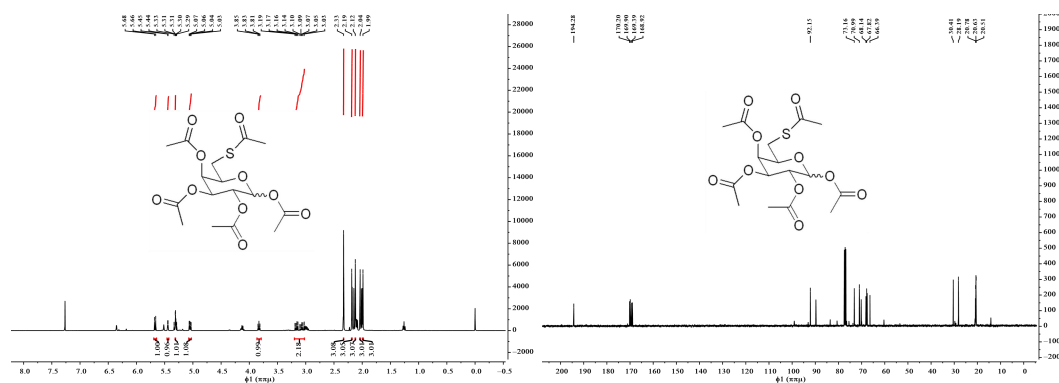
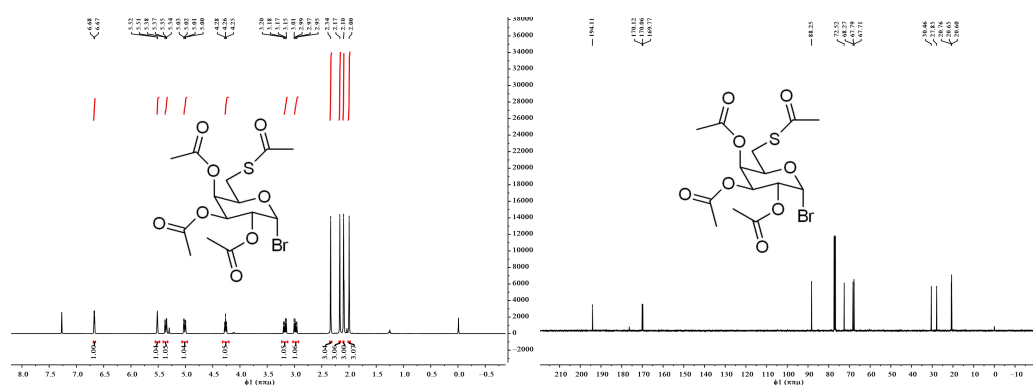


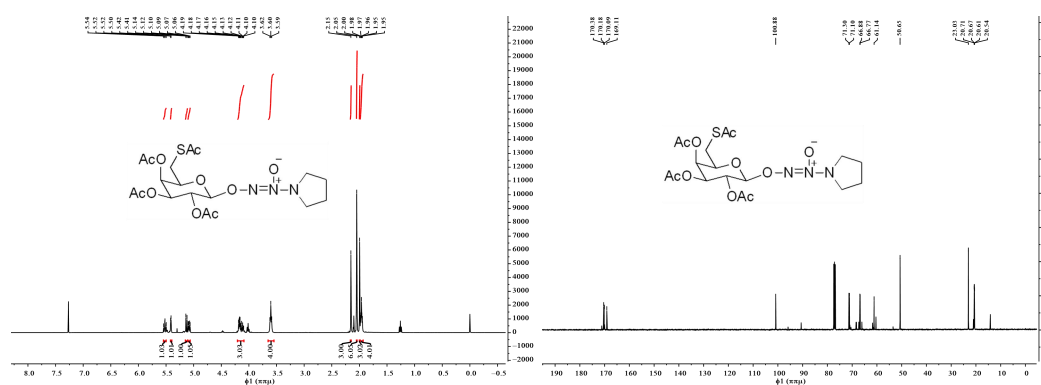
Figure S3.  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) spectrum of compound 3.



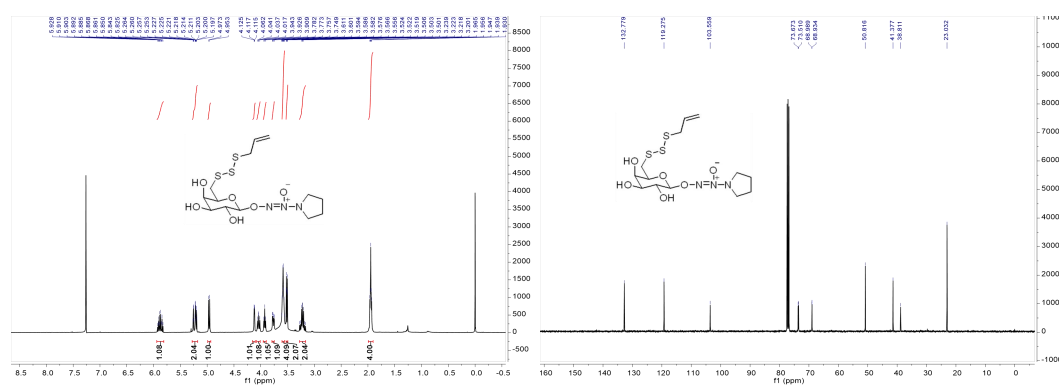
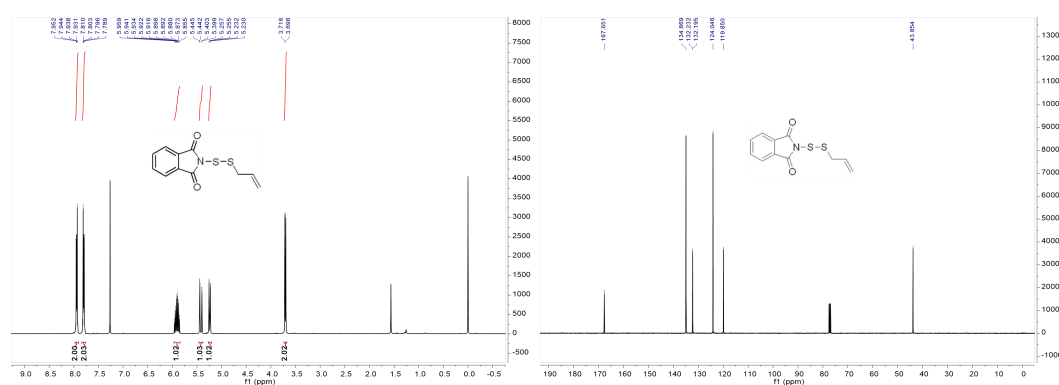
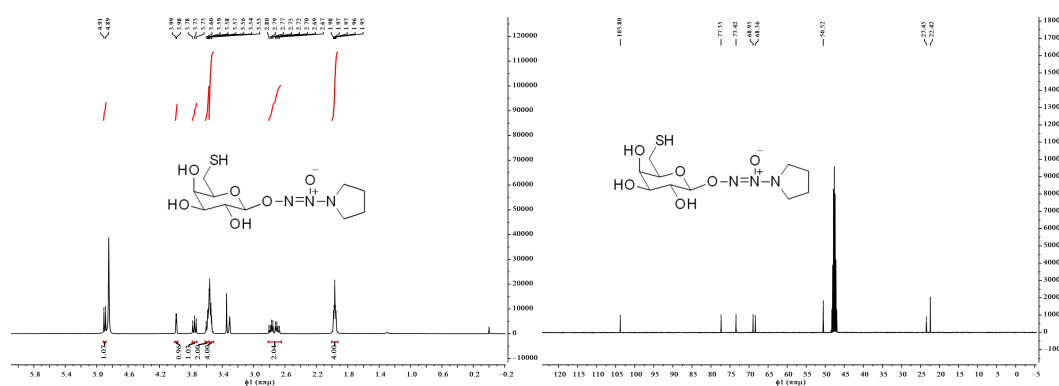
**Figure S4.**  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) spectrum of compound 5.

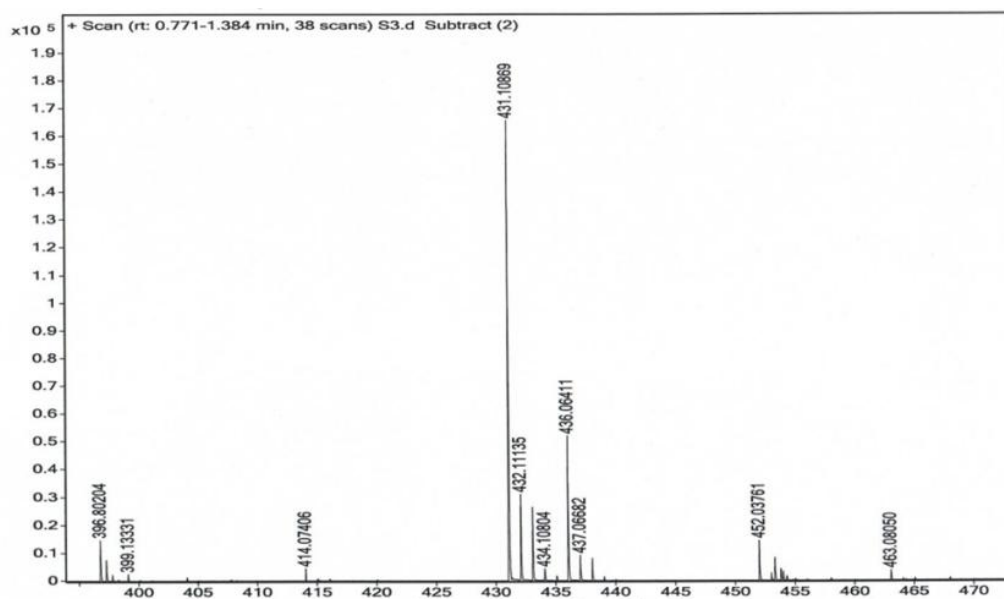


**Figure S5.**  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) spectrum of compound 6.

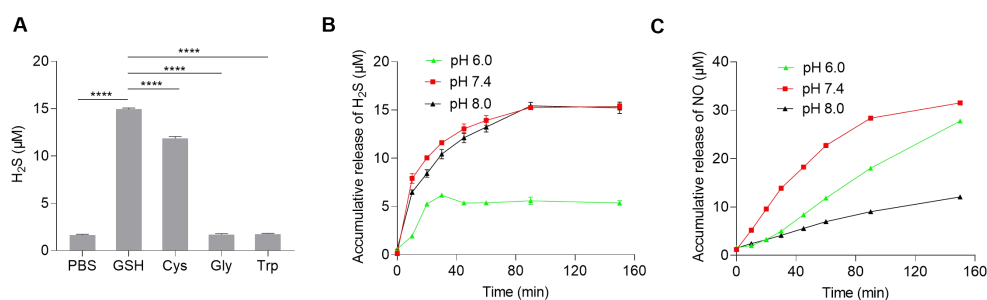


**Figure S6.**  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) spectrum of compound 7.

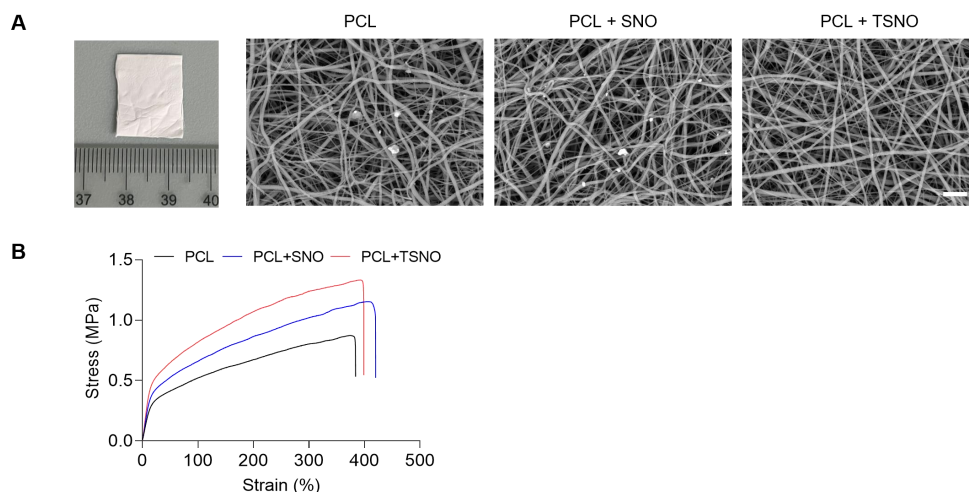




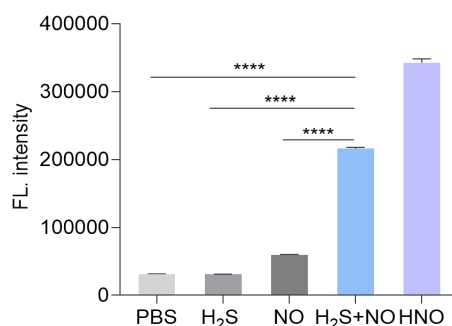
**Figure S9.**  $^1\text{H}$  NMR (left) and  $^{13}\text{C}$  NMR (right) of compound TSNO (up). HRMS spectrum of compound TSNO (down).



**Figure S10.** *In vitro* release of  $\text{H}_2\text{S}$  and NO from the TSNO prodrug. (A)  $\text{H}_2\text{S}$  release from TSNO with various thiol catalysts. (B) *In vitro* release of  $\text{H}_2\text{S}$  from the TSNO prodrug in the presence of GSH (100  $\mu\text{M}$ ) at various pH values ( $n=3$ ). (C) *In vitro* release of NO from the TSNO prodrug in the presence of GSH (100  $\mu\text{M}$ ) and engineered A4- $\beta$ -Gal $^{\text{H}363\text{A}}$  (0.005 mg/mL) at various pH values ( $n=3$ ).



**Figure S11.** The characterization of TSNO-functionalized wound dressing. (A) Representative images of the wound dressing and its microstructure demonstrated by SEM (scale bar, 100  $\mu\text{m}$ ). (B) Characterization of the wound dressing in terms of mechanical properties.



**Figure S12.** HNO production from the interaction of H<sub>2</sub>S and NO. An HNO-specific fluorescence probe (10  $\mu\text{M}$ ) was co-incubated with the TSNO prodrug in the presence of GSH (100  $\mu\text{M}$ ) and engineered A4- $\beta\text{-Gal}^{\text{H363A}}$  (0.005 mg/mL). Angeli's salt (an HNO donor) was used as a positive control, excitation wavelength 360 nm, emission wavelength 460 nm (n=3).

**Table S1.** Primer sequences of qPCR.

Gene name	Forward primer (5' to 3')	Reverse primer (5' to 3')
m-Gapdh	CCCTTATTGACCTCAACTACA	TGGTGAGGGGCCATCCACAGTCTTCTG
m-Pecam1	ACGCTGGTGCTCTATGCAAG	TCAGTTGCTGCCCATTCATCA
m-Angpt1	CACATAGGGTGCAGCAACCA	CGTCGTGTTCTGGAAGAATGA
m-Vegfa	CTGCCGTCGATTGAGACC	CCCCTCCTTGTAACCACTGTC
m-Kdr	TTTGCAAATACAACCCTTCAGA	GCAGAAGATACTGTCAACCACC

m-Mrc1	CTCTGTTTCAGCTATTGGACGC	CGGAATTTCTGGGATTCAGCTTC
m-Arg1	CTCCAAGCCAAAGTCCTTAGAG	AGGAGCTGT CATTAGGGACATC
m-Il4	ATCCTGCTCTTCTTTCTCGAATGT	GCCGATGATCTCTCTCAAGTGATT
m-Il10	AGCCTTATCGGAAATGATCCAGT	GGCCTTGTAGACACCTTGGT
m-Nos2	GTTCTCAGCCCAACAATACAAGA	GTGGACGGGTCGATGTCAC
m-Il1b	AGCTCTCCACCTCAATGGAC	GACAGGCTTGTGCTCTGCTT
m-Il6	TCCATCCAGTTGCCTTCTTG	GGTCTGTTGGGAGTGGTATC
m-Tnf	ACGGCATGGATCTCAAAGAC	CGGACTCCGCAAAGTCTAAG

---