Supplementary Materials

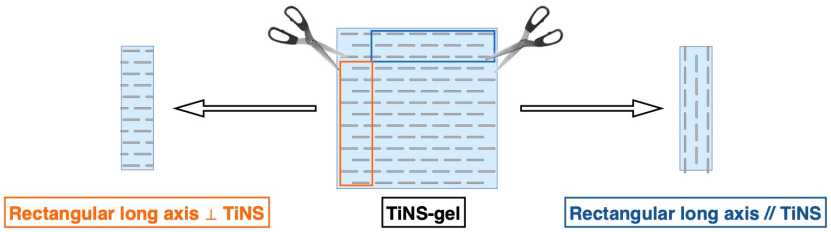

Figure S1. Schematic illustration of the trimming of TiNS-gel into two types of rectangular strips in such a way that the rectangular long axis become perpendicular (left) and parallel (right) to the alignment of TiNS platelets.

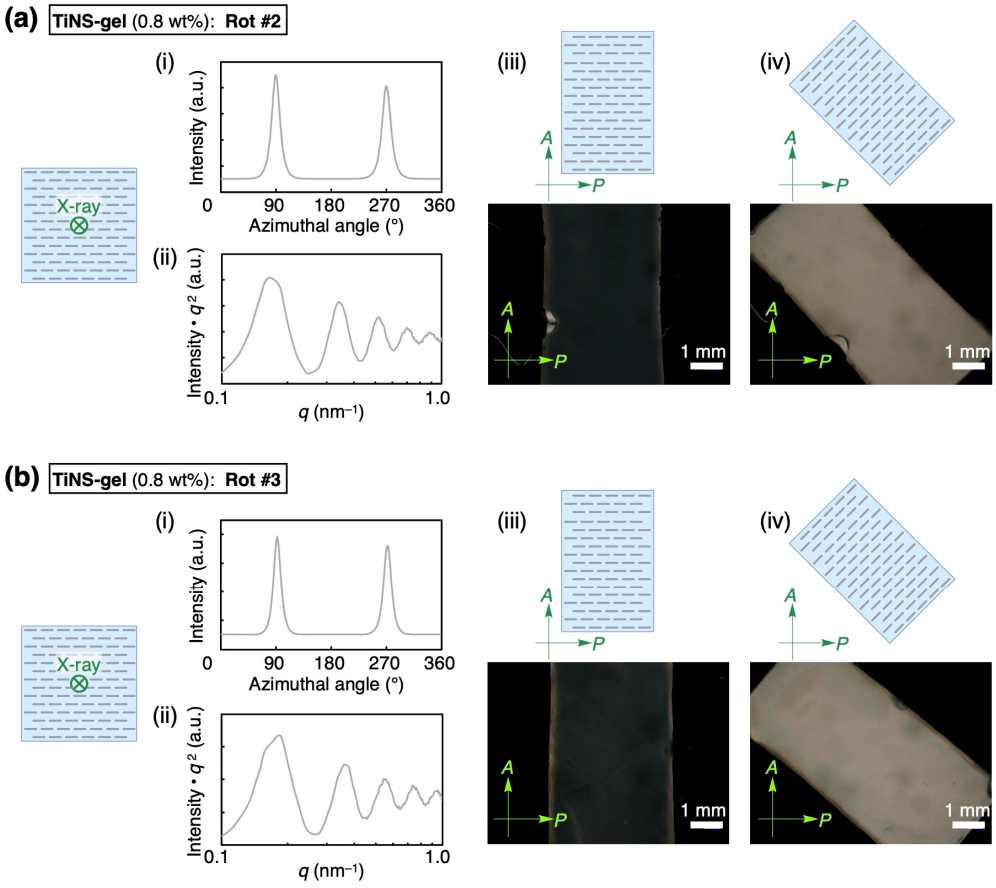

Figure S2. (a,b) Reproducibility of the synthesis of TiNS-gel. In the same procedure similar to Figure 1, two more samples of TiNS-gel at [TiNS] = 0.8 wt% were synthesized (a, Rot #2; b, Rot #3) and characterized by (i, ii) SAXS and (iii, iv) POM.

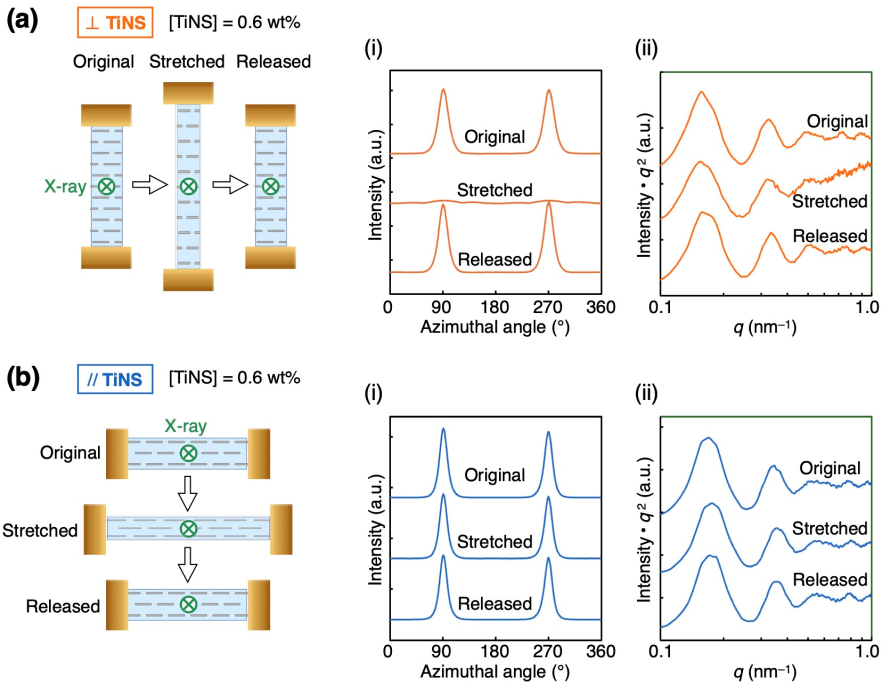

Figure S3. (a,b) Changes in the SAXS profiles of TiNS-gel ([TiNS] = 0.6 wt%) upon tensile deformation up to 25% strain along parallel (a) and vertical (b) to the alignment of TiNS platelets. Measurements were taken before deformation (original), after deformation (stretched), and then after releasing the force (released): (i) intensity–azimuthal angle plots and (ii) intensity–*q* plots.

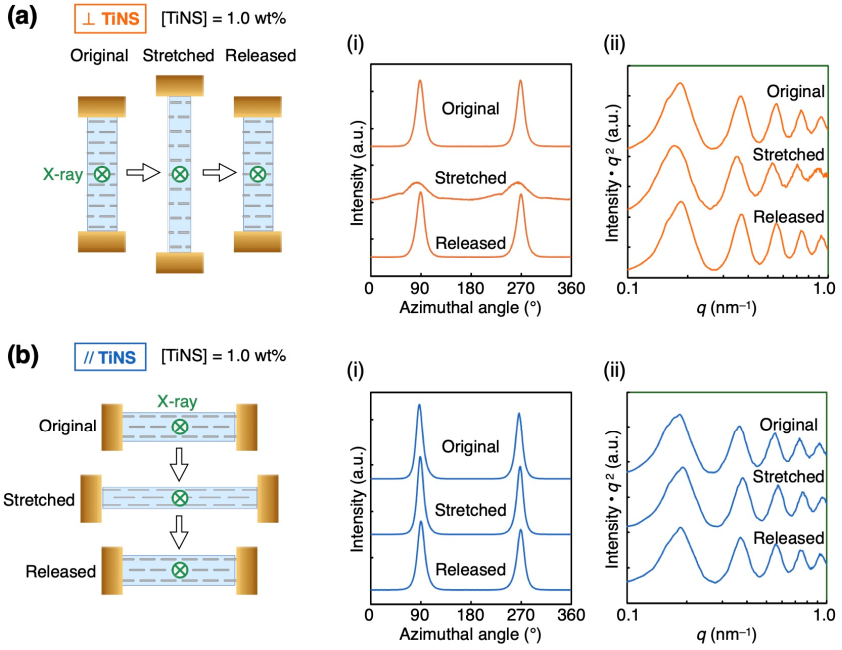

Figure S4. (a,b) Changes in the SAXS profiles of TiNS-gel ([TiNS] = 1.0 wt%) upon tensile deformation up to 25% strain along parallel (a) and vertical (b) to the alignment of TiNS

platelets. Measurements were taken before deformation (original), after deformation (stretched), and then after releasing the force (released): (i) intensity–azimuthal angle plots and (ii) intensity–*q* plots.

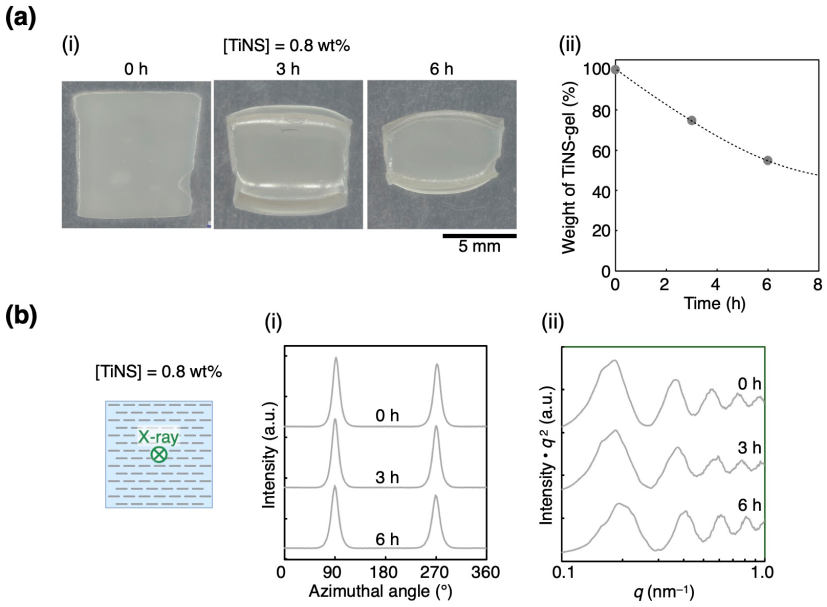

Figure S5. (a) Drying profiles of TiNS-gel ([TiNS] = 0.8 wt%) in open air (25 °C, 50% RH) for 6 hours. Changes in (i) size and (ii) weight. (b) Changes in the SAXS profiles of TiNS-gel during the drying process: (i) intensity–azimuthal angle plots and (ii) intensity–*q* plots.

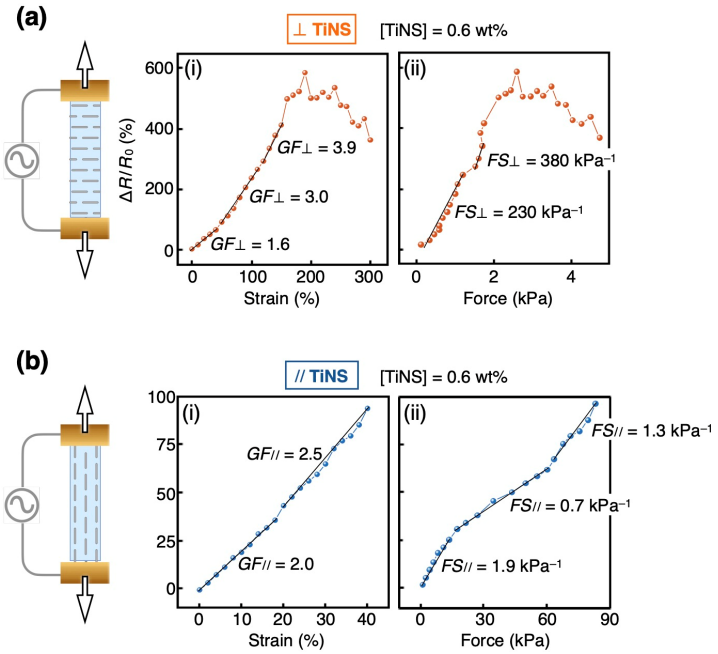

Figure S6. (a,b) Relative resistance change ($\Delta R / R_0$) of TiNS-gel ([TiNS] = 0.6 wt%) upon tensile deformation along parallel (a) and vertical (b) to the alignment of TiNS platelets: plots

of (i) $\Delta R / R_0$ versus strain and (ii) $\Delta R / R_0$ versus force.

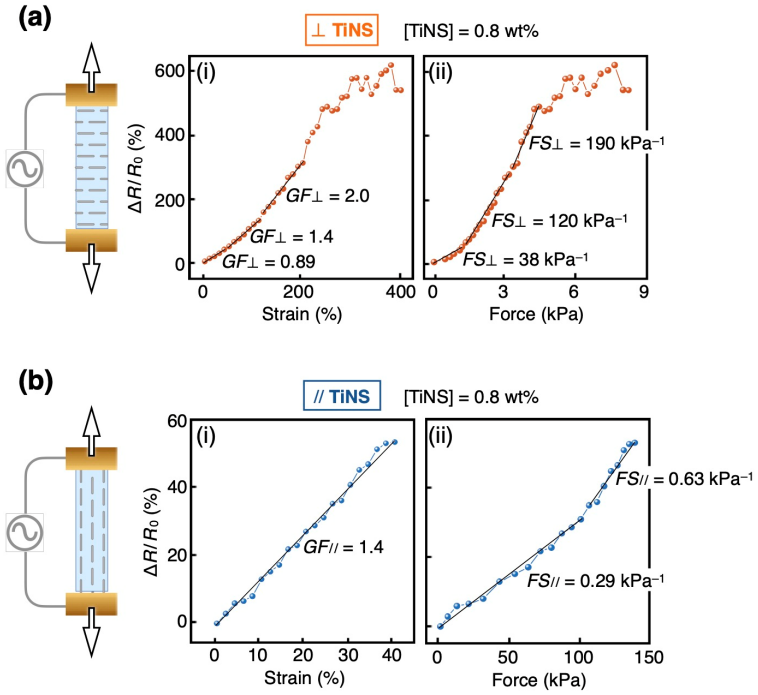

Figure S7. (a,b) Relative resistance change ($\Delta R / R_0$) of TiNS-gel ([TiNS] = 0.8 wt%) upon tensile deformation along parallel (a) and vertical (b) to the alignment of TiNS platelets: plots of (i) $\Delta R / R_0$ versus strain and (ii) $\Delta R / R_0$ versus force.

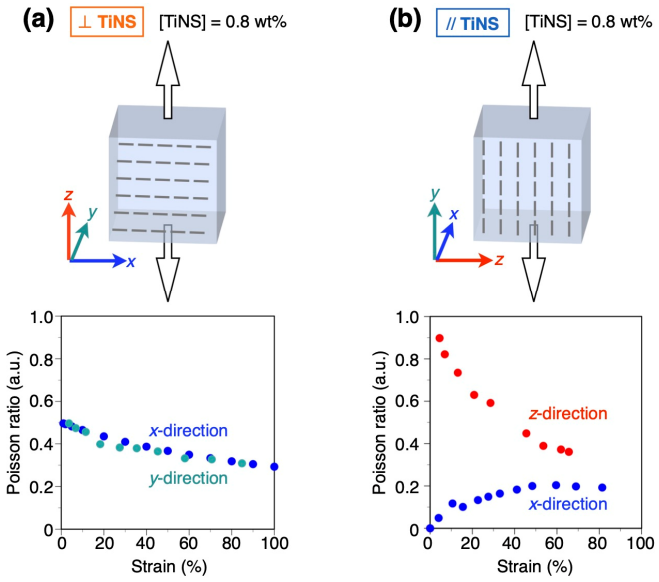

Figure S8. (a,b) Changes in the Poisson ratios of TiNS-gel ([TiNS] = 0.8 wt%) upon tensile deformation along parallel (a) and vertical (b) to the alignment of TiNS platelets.