

## Supporting information

### Intergrowth between the Oxynitride Perovskite $\text{SrTaO}_2\text{N}$ and a Ruddlesden-Popper Phase $\text{Sr}_2\text{TaO}_3\text{N}$

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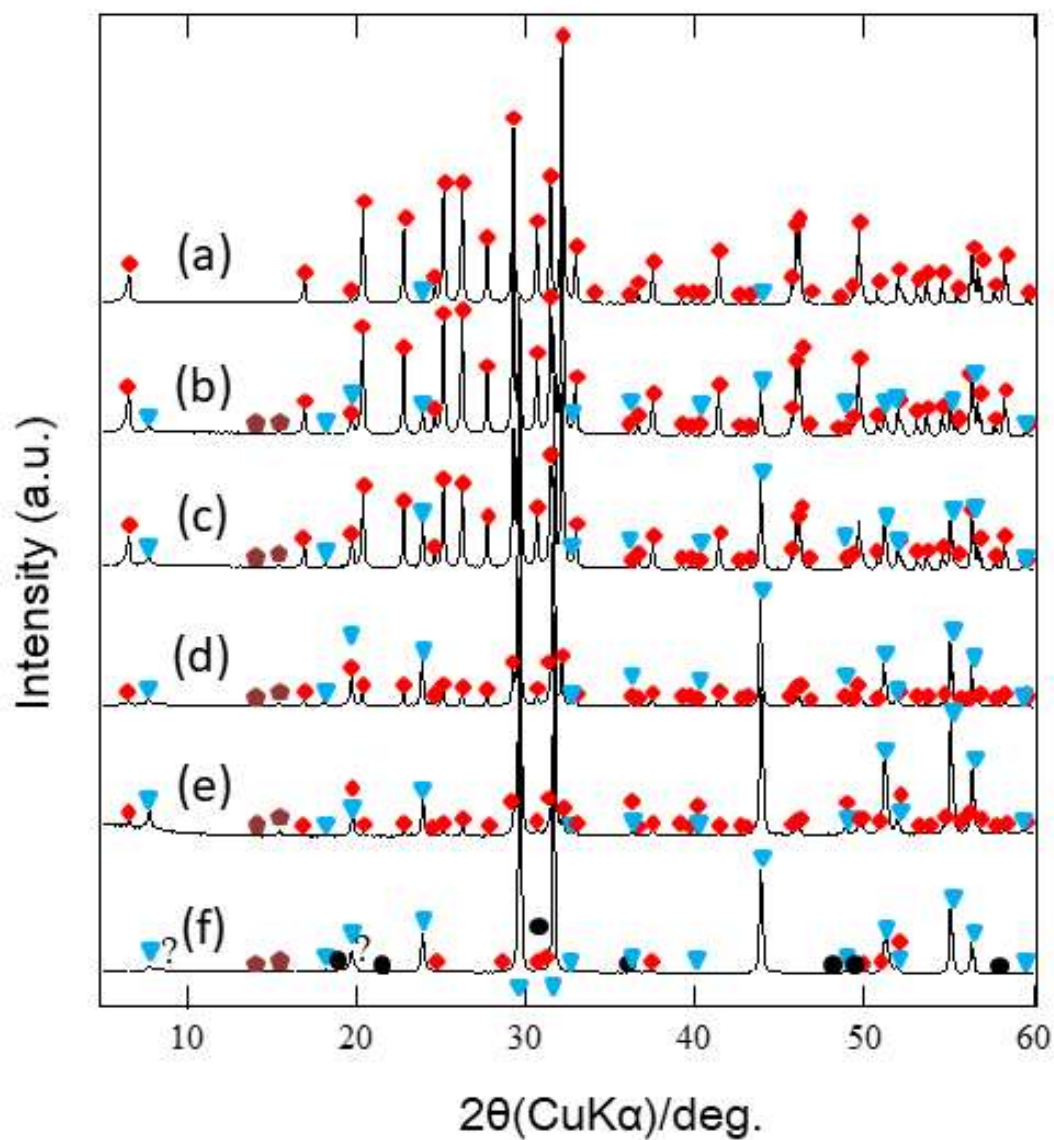


Fig. S1 Powder X-ray diffraction patterns for oxide precursors (a) prepared with  $x = 1.0$ , (b)  $x = 1.05$ , (c)  $x = 1.1$ , (d)  $x = 1.2$ , (e)  $x = 1.25$  and (f)  $x = 1.3$ . Red squares, blue triangles, black circles and brown pentagons indicate diffraction peaks attributed to  $\text{Sr}_2\text{Ta}_2\text{O}_7$ ,  $\text{Sr}_5\text{Ta}_4\text{O}_{15}$ ,  $\text{Sr}_{1.4}\text{Ta}_{0.6}\text{O}_{2.7}$  and  $\text{Sr}(\text{OH})_2(\text{H}_2\text{O})_8$ , respectively. Weak diffraction peaks marked by question marks are not assigned. Sr/Ta atomic ratios determined by XRF were (a) 0.98(2), (b) 1.03(2), (c) 1.08(2), (d) 1.17(3), (e) 1.22(2) and (f) 1.27(3), respectively.

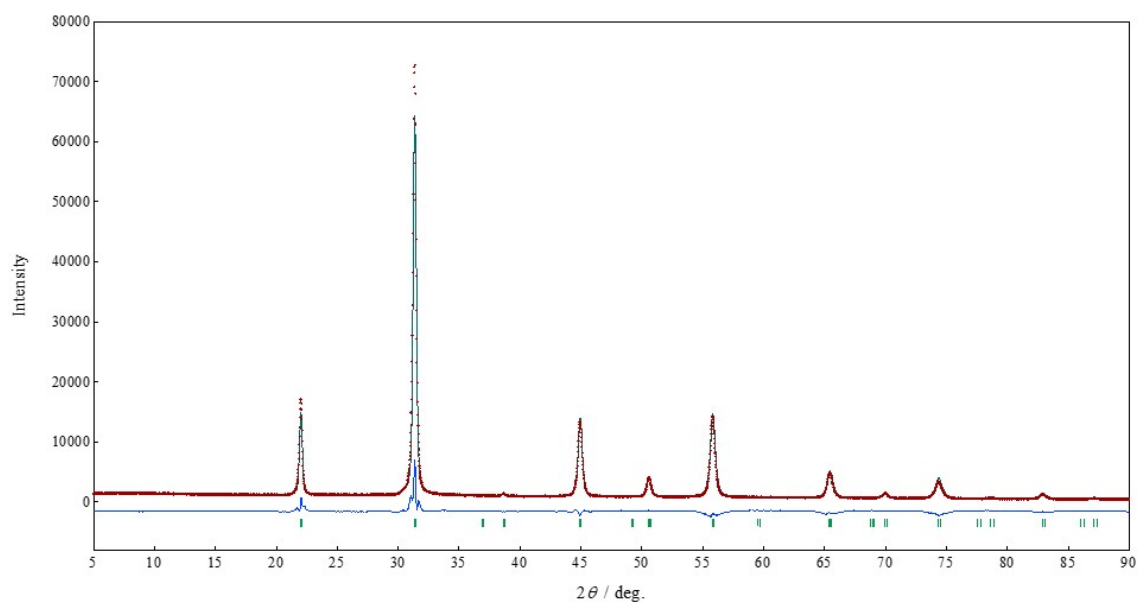


Fig. S2 Observed (red) and calculated (green)XRD patterns for SrTaO<sub>2</sub>N. Their difference plot is shown in blue. Green ticks show the peak positions expected for SrTaO<sub>2</sub>N in the  $I4/mcm$  space group.

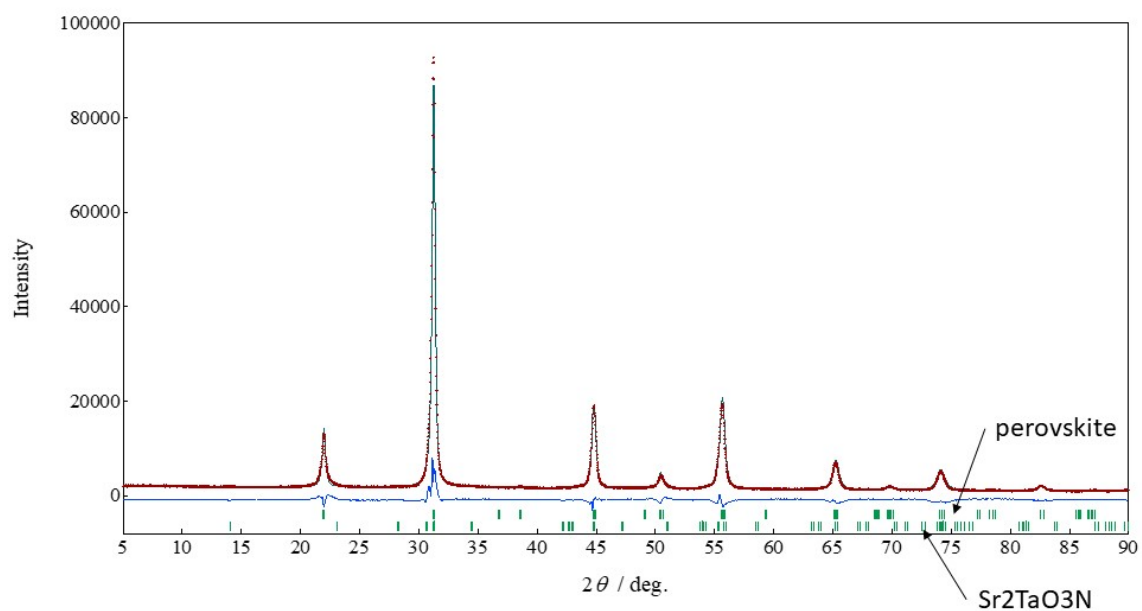


Fig. S3 Observed (red) and calculated (green) XRD patterns for  $\text{Sr}_{1.2}\text{TaO}_2\text{N}$ . Their difference plot is shown in blue. Green ticks above and below show the peak positions expected for  $\text{SrTaO}_2\text{N}$  perovskite in the  $I4/mcm$  space group and  $\text{Sr}_2\text{TaO}_3\text{N}$  RP phase in the  $I4/mmm$  space group, respectively.

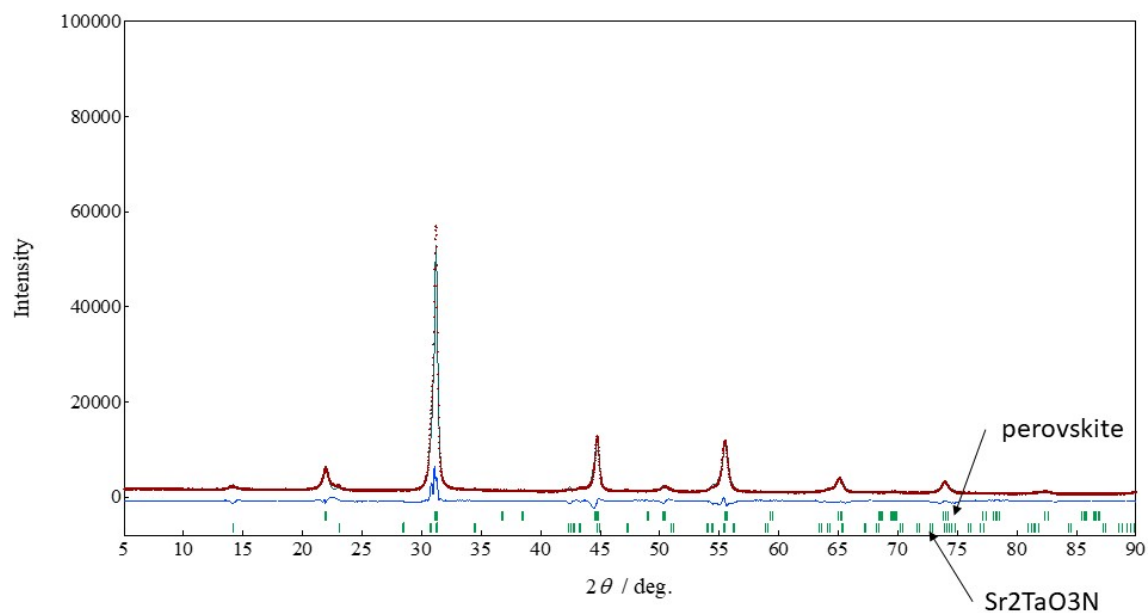


Fig. S4 Observed (red) and calculated (green) XRD patterns for  $\text{Sr}_{1.4}\text{TaO}_2\text{N}$ . Their difference plot is shown in blue. Green ticks above and below show the peak positions expected for  $\text{SrTaO}_2\text{N}$  perovskite in the  $I4/mcm$  space group and  $\text{Sr}_2\text{TaO}_3\text{N}$  RP phase in the  $I4/mmm$  space group, respectively.

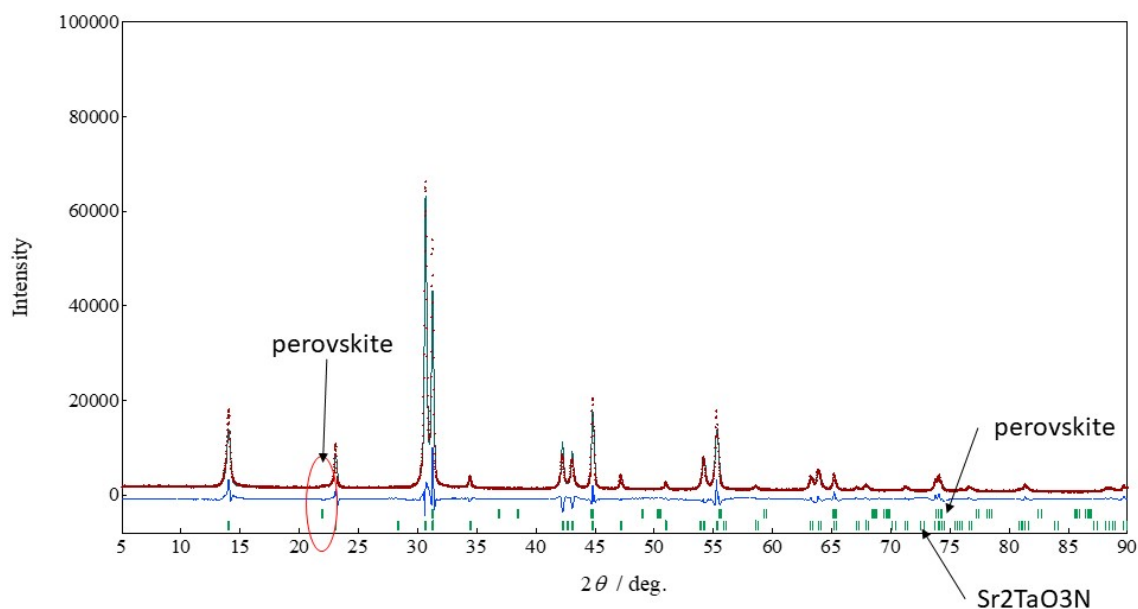


Fig. S5 Observed (red) and calculated (green) XRD patterns for  $\text{Sr}_2\text{TaO}_3\text{N}$ . Their difference plot is shown in blue. Green ticks above and below show the peak positions expected for  $\text{SrTaO}_2\text{N}$  perovskite in the  $I4/mcm$  space group and  $\text{Sr}_2\text{TaO}_3\text{N}$  RP phase in the  $I4/mmm$  space group, respectively. Presence of perovskite is not so apparent even from the diffraction circled in red.

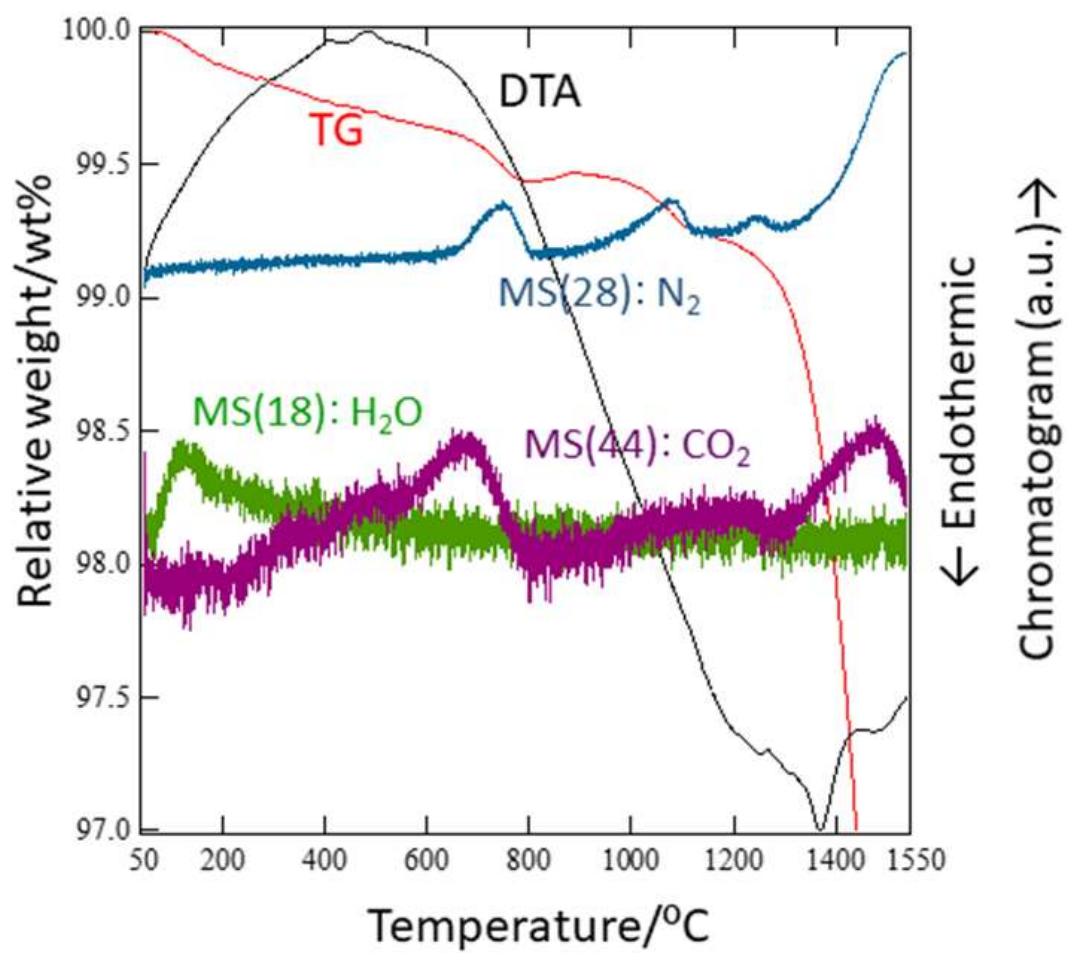


Fig. S6 TG-DTA-MS data acquired from  $\text{Sr}_2\text{TaO}_3\text{N}$  in a helium atmosphere.