

Supplementary Information

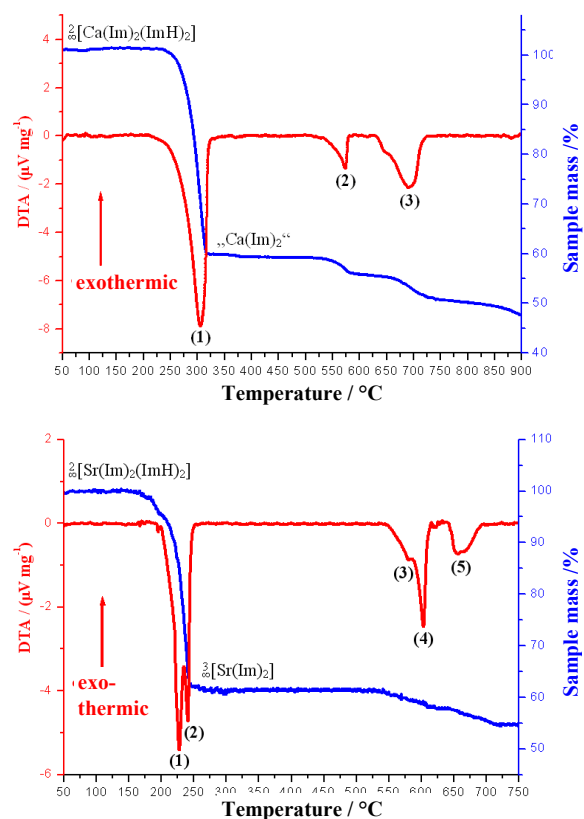


Fig. S1 Thermal analyses investigated by simultaneous DTA/TG for $2[\text{Ca}(\text{Im})_2(\text{ImH})_2]$ (2) in the temperature range of 20 to 900 °C (top), and for $2[\text{Sr}(\text{Im})_2(\text{ImH})_2]$ (3) in the temperature range of 20 to 750 °C (bottom) with a heating rate of 10 °C/min in a continuous Ar flow of 60 mL/min.

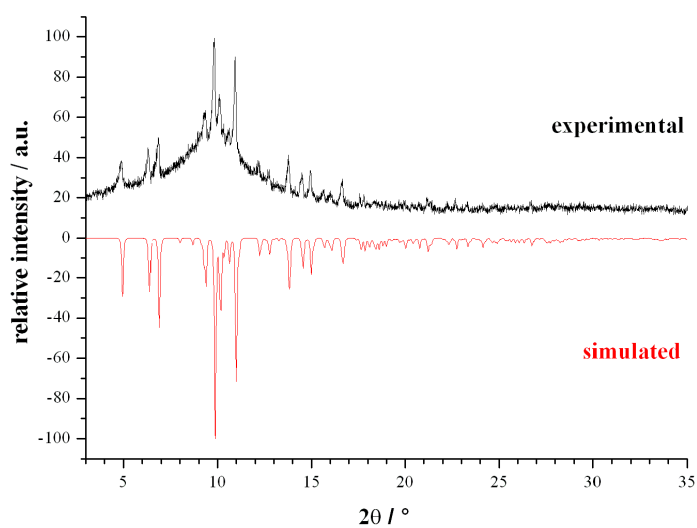


Fig. S2 Comparison of the X-Ray powder pattern of $2[\text{Ca}(\text{Im})_2(\text{ImH})_2]$ (2) with a simulated pattern based on the single crystal X-ray refinement indicating absorption as well as phase purity of the compound.

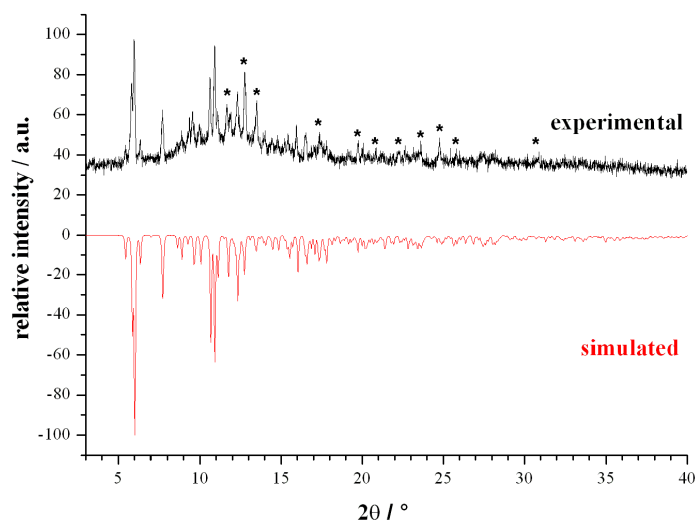


Fig. S3 Comparison of the X-Ray powder pattern of $[\text{Sr}(\text{Im})_2(\text{ImH})_2]$ (**3**) with a simulated pattern based on the single crystal X-ray refinement indicating some absorption as well as phase impurity of SrH_2 . Reflections marked with an asterisk derive from or partly contain intensity of the hydride.

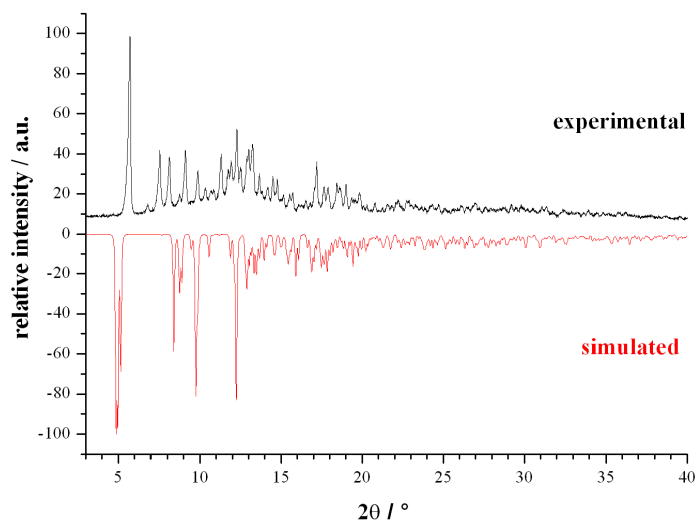


Fig. S4 Comparison of the X-Ray powder pattern of the bulk phase with the constitution $[\text{BaIm}]_2$ with a simulation of a powder pattern of $[\text{Ba}(\text{Im})_2(\text{ImH})_2]$ (**4**) based on the single crystal X-ray refinement clearly shows that the strand structure **4** is not present in the bulk material.

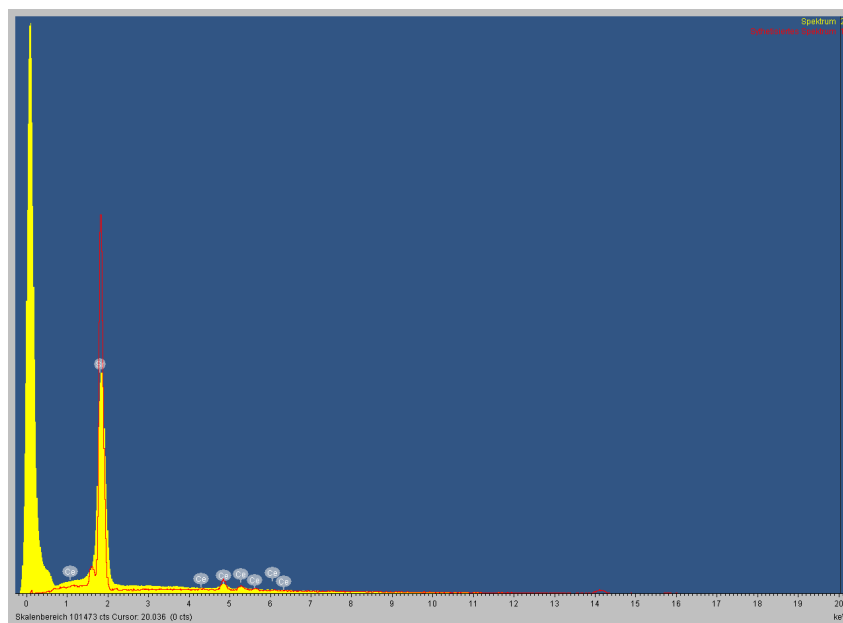


Fig. S5 Elemental analysis of the strontium and cerium content of ${}^2_{\infty}[\text{Sr}(\text{Im})_2(\text{ImH})_2]:\text{Ce}^{3+}$ (**3**:**Ce**³⁺) by electron dispersive X-ray absorption spectroscopy giving a ratio of 94:6 (yellow) in comparison to the starting ratio of 90:10 (red).

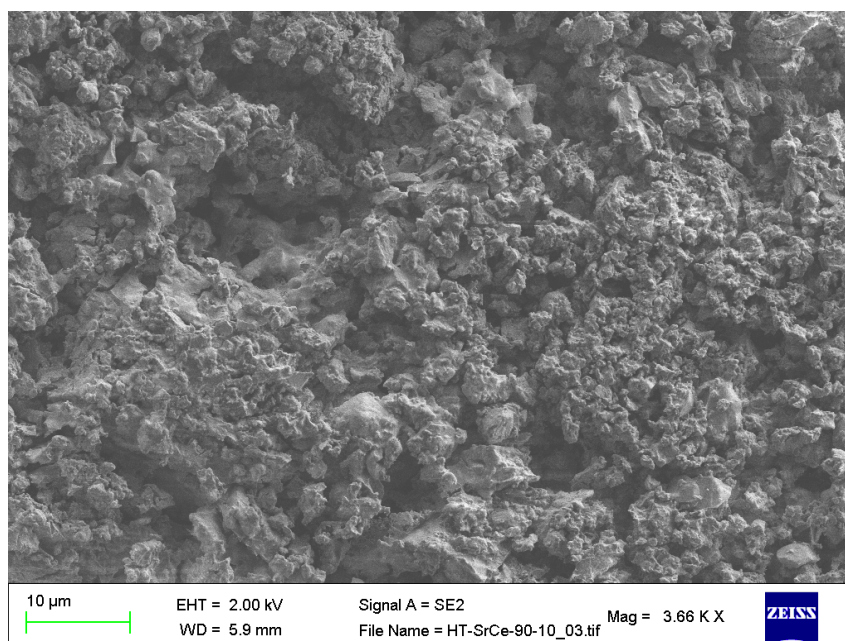


Fig. S6 SEM image of the microcrystalline sample of ${}^2_{\infty}[\text{Sr}(\text{Im})_2(\text{ImH})_2]:\text{Ce}^{3+}$ (**3**:**Ce**³⁺) used for the electron dispersive X-ray absorption spectroscopy investigation.

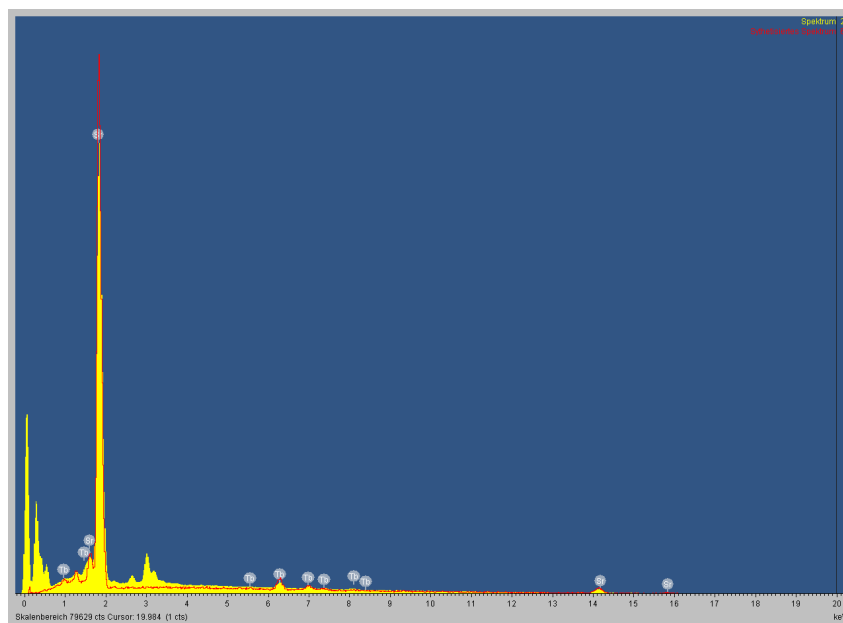


Fig. S7 Elemental analysis of the strontium and terbium content of ${}^2_{\infty}[\text{Sr}(\text{Im})_2]\cdot\text{Tb}^{3+}$ (**5:Tb³⁺**) by electron dispersive X-ray absorption spectroscopy giving a ratio of 96:4 (yellow) in comparison to the starting ratio of 90:10 (red).

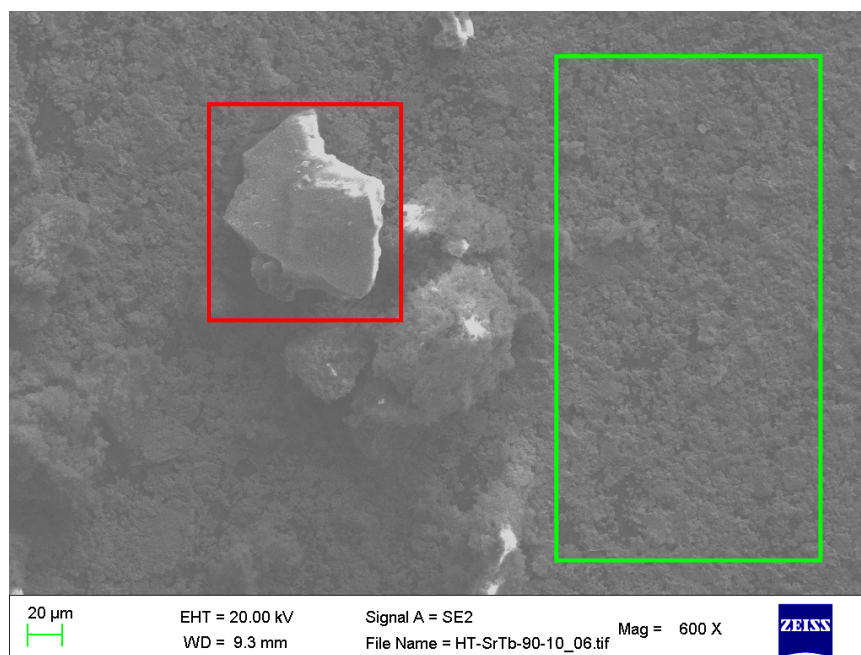


Fig. S8 SEM image of the microcrystalline sample of ${}^2_{\infty}[\text{Sr}(\text{Im})_2]\cdot\text{Tb}^{3+}$ (**5:Tb³⁺**) used for the electron dispersive X-ray absorption spectroscopy investigation on the bulk material with a 0.04Tb³⁺ doping (green frame) and the unknown minority side phase (red frame).