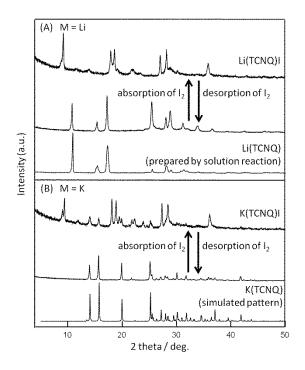
## Supporting Information

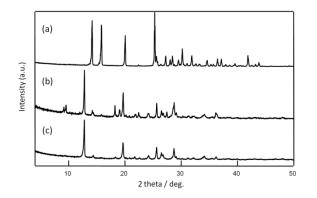
## **Reversible iodine adsorption by alkali-TCNQ salts with associated changes in physical properties**

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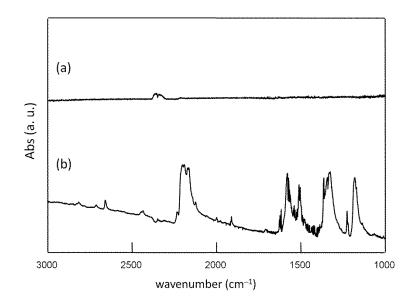
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**Fig. S1.** Changes of the powder XRD patterns of M(TCNQ) caused by reversible iodine absorption for the salts: (A) Li and (B) K.



**Fig. S2.** Powder XRD patterns of (a) K(TCNQ) prepared by solution reactions, (b) an over-doped salt obtained by storing K(TCNQ) in an iodine atmosphere for one month, which contains a small portion of K(TCNQ)I, and (c) the over-doped salt obtained by stirring a dispersion of K(TCNQ) in a hexane solution of iodine for two days.



**Fig. S3.** IR spectra of (a) Na(TCNQ)I prepared by solid-state reactions and (b) Na(TCNQ) obtained by annealing Na(TCNQ)I.

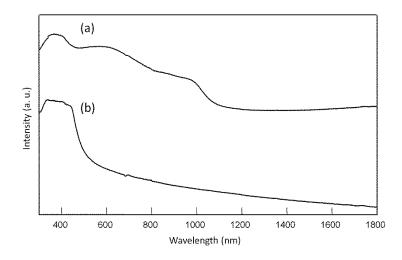


Fig. S4. Solid-state UV-vis-NIR absorption spectra of (a)  $Rb_2(TCNQ)_3I_2$  prepared by solid-state reaction and (b) TCNQ.

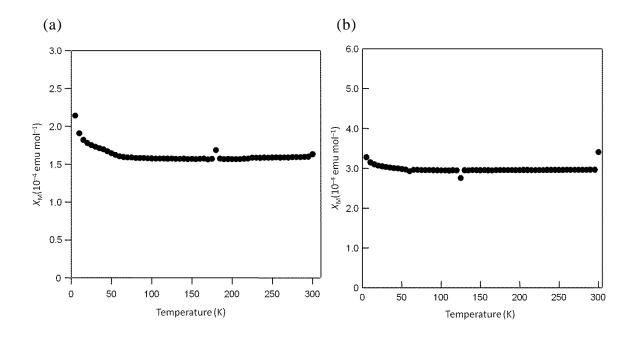


Fig. S5. Temperature dependence of the magnetic susceptibilities of (a)  $Na(TCNQ)I_{6.0}$  and (b)  $K(TCNQ)I_{5.8}$  prepared by liquid phase reactions.

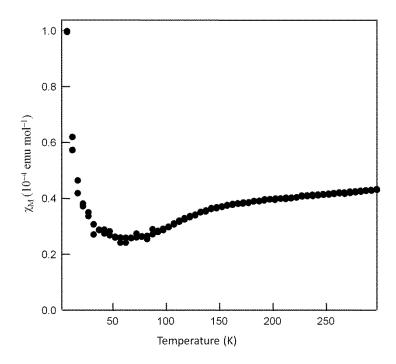


Fig. S6. Temperature dependence of the magnetic susceptibility of  $Rb_2(TCNQ)_3I_2$  prepared by solid-state reaction.