Supporting Information

2-D Hydrogen-bonded Square-grid Coordination Networks with a Substitution-active Metal Site

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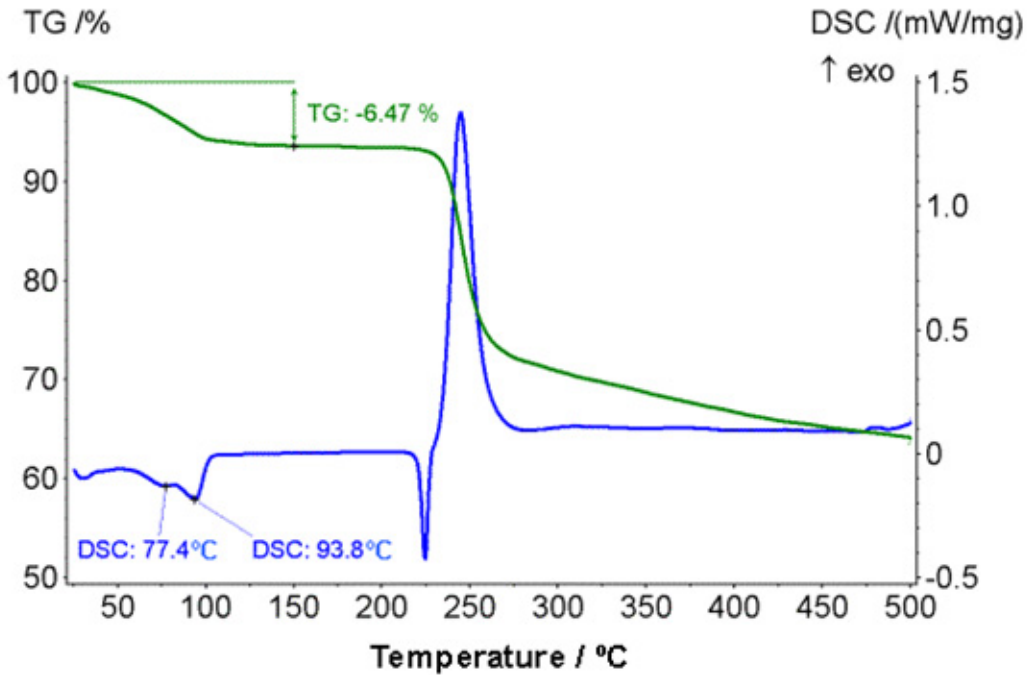

Figure S1. TG/DSC curves of **2** measured in N_2 (heating rate is 2 °C /min)

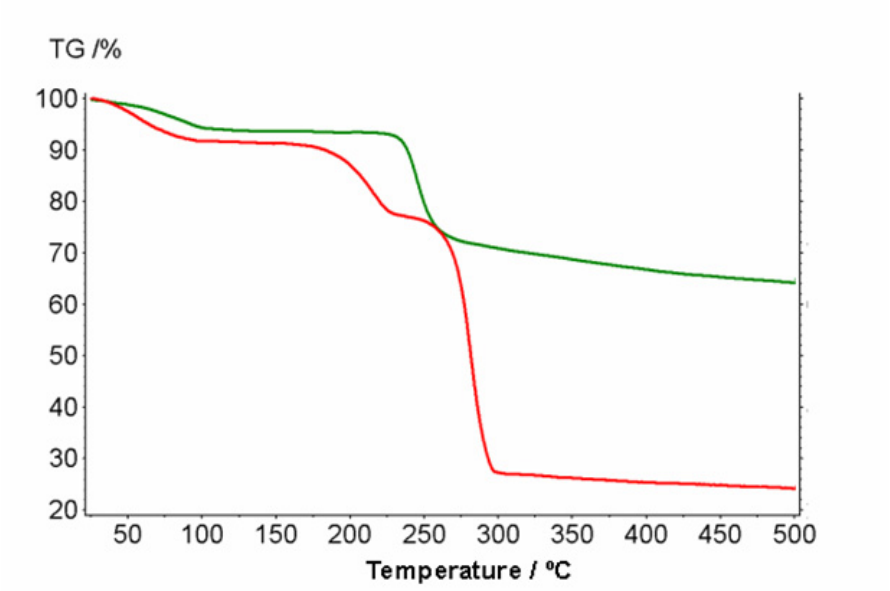

Figure S2. TG curve of **2** (green) and **5** (red) measured in N_2 (heating rate is 2 °C /min)

Comparison by thermogravimetric analysis: As shown in Figure S2, **2** showed much higher thermal stability than **5** (decomposition temperature: **2**, 220 °C; **5**, 170 °C).

Comparison by X-ray analysis: Whereas **2** kept its crystallinity even at high temperature (<150 °C), crystal **5** gradually deteriorated above room temperature and no longer diffracted at 60 °C.

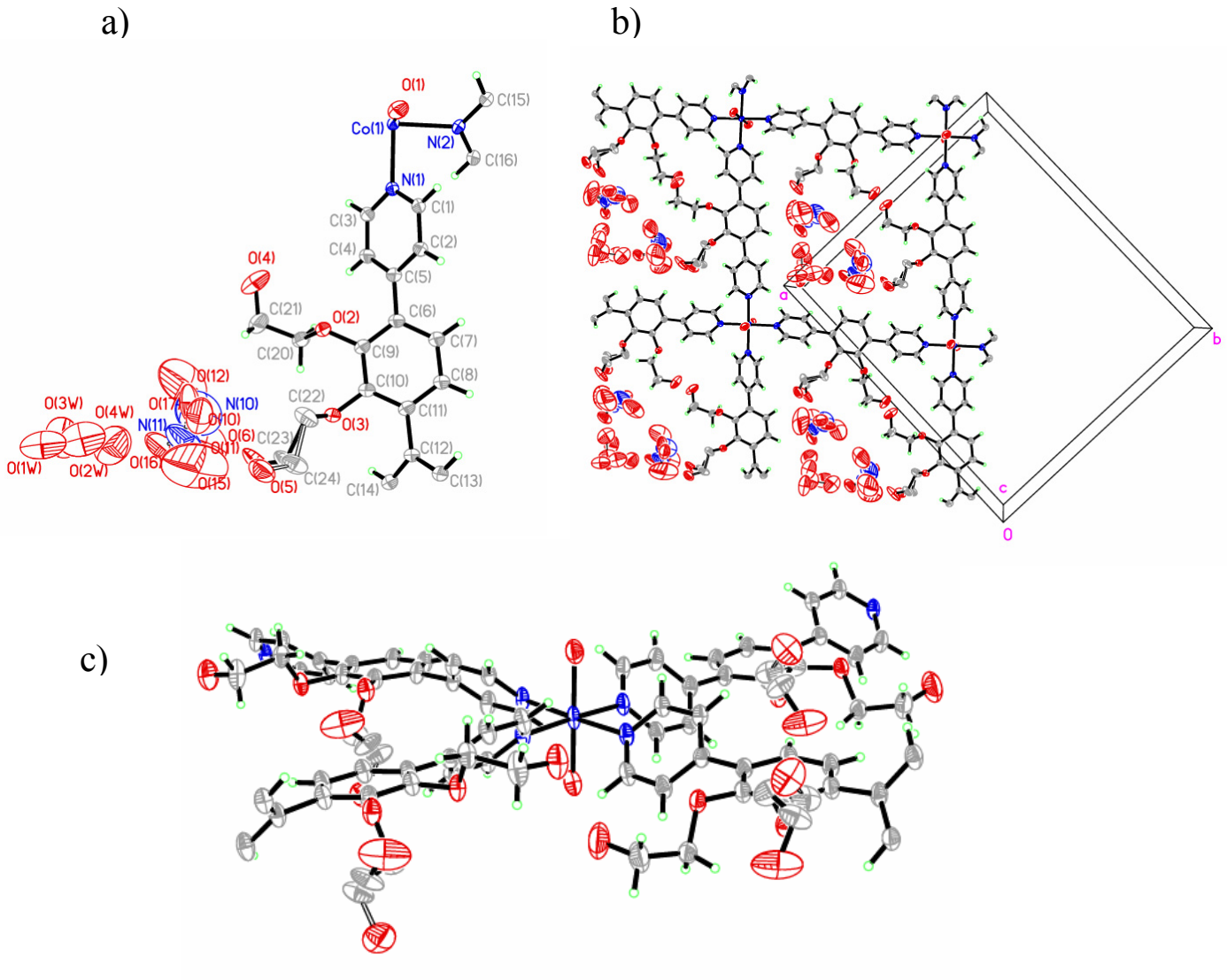

Figure S3. ORTEP view (50% probability level) of **2** (80 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Co(II) atom. The thermal temperature factors of all non-hydrogen atoms were anisotropically refined. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered ethylene glycol groups, nitrate ions and water molecules.

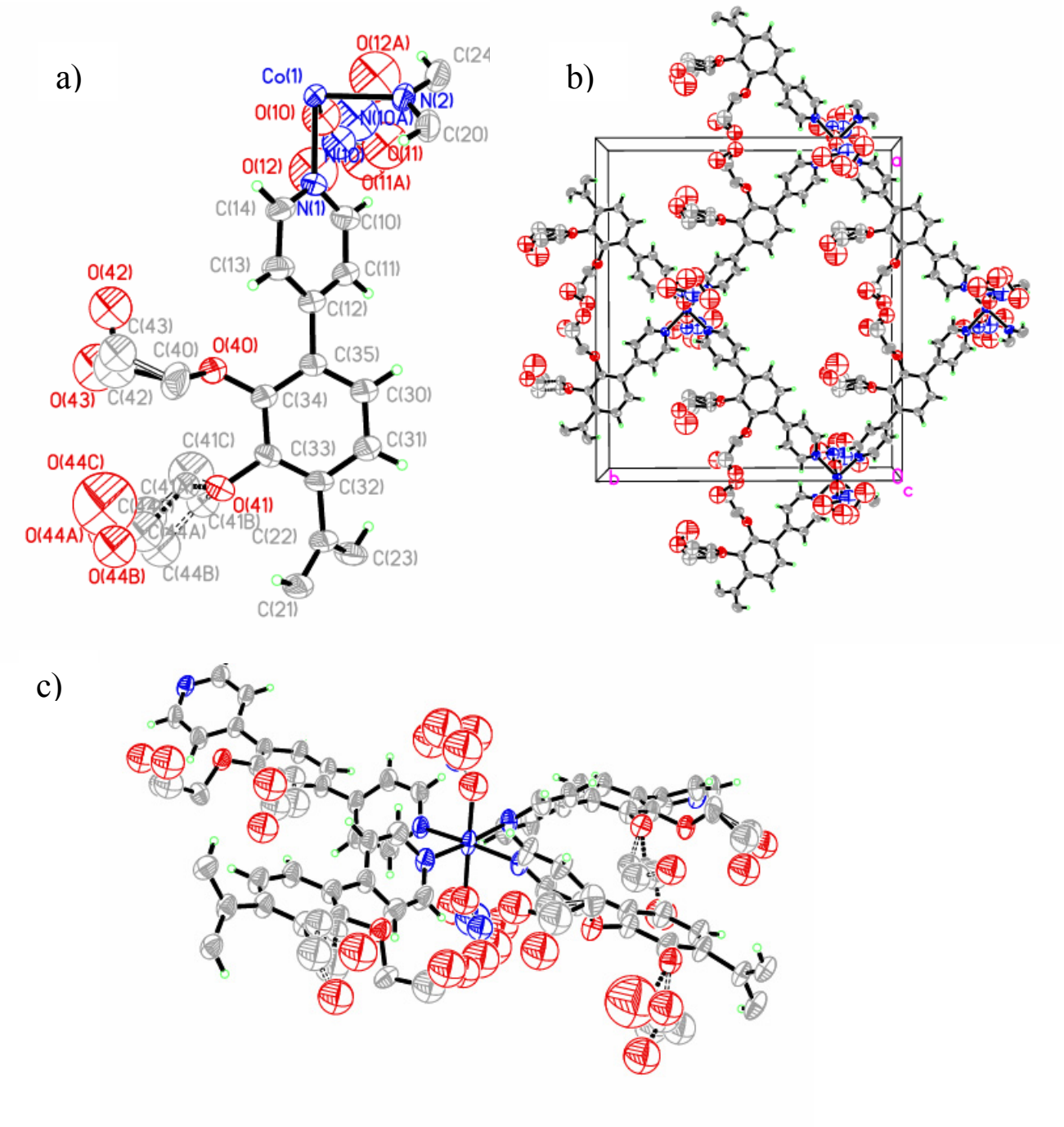

Figure S4. ORTEP view (50% probability level) of **6** (423 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Co(II) atom. The thermal temperature factors of nitrate ions and ethylene glycol groups were isotropically refined because of severe disorder. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered nitrate ions and disordered ethylene glycol groups.

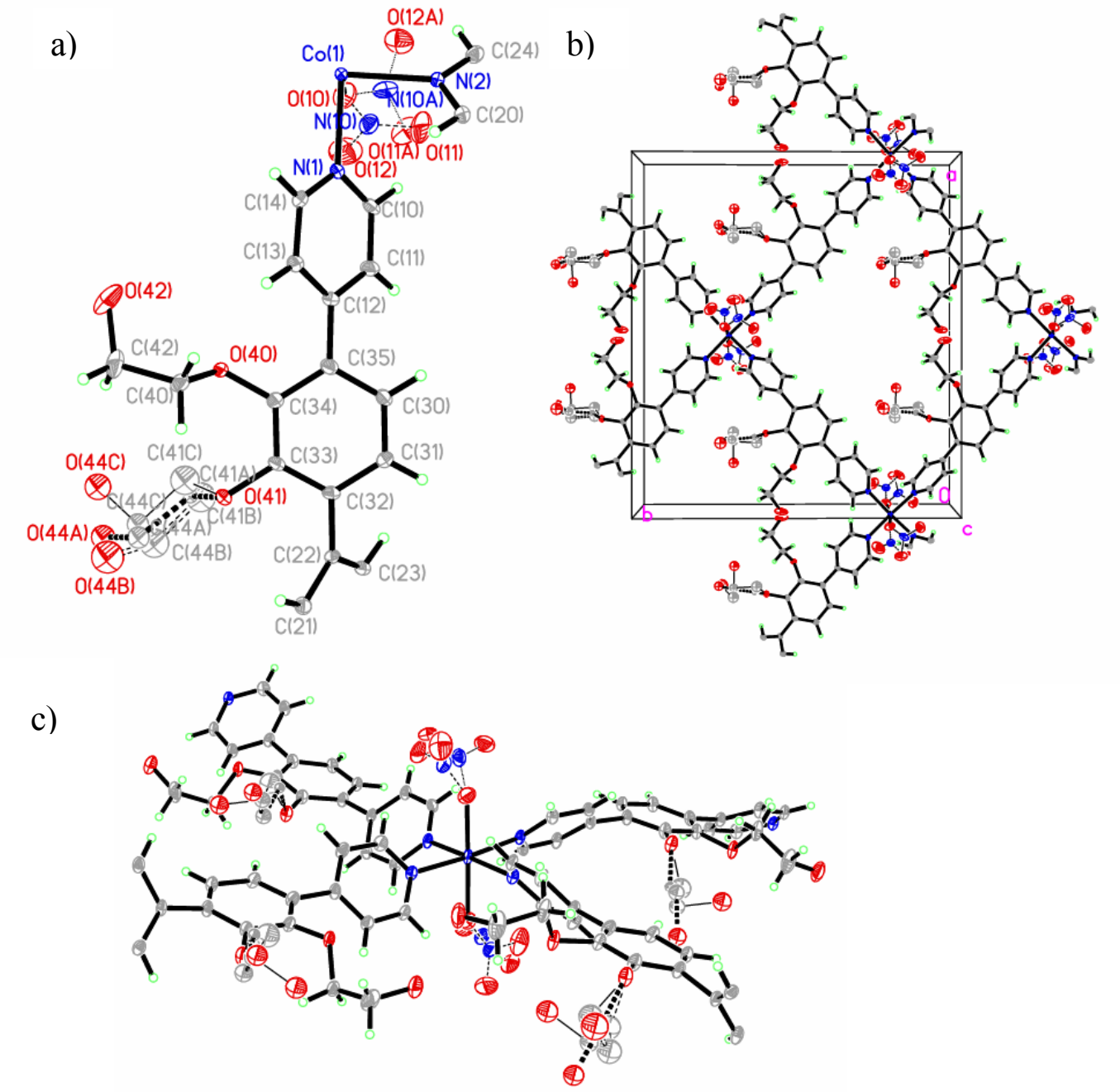

Figure S5. ORTEP vie w (50% probability level) of **6** (80 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Co(II) atom. The thermal temperature factors of ethylene glycol groups were isotropically refined because of severe disorder. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered nitrate ions and ethylene glycol groups.

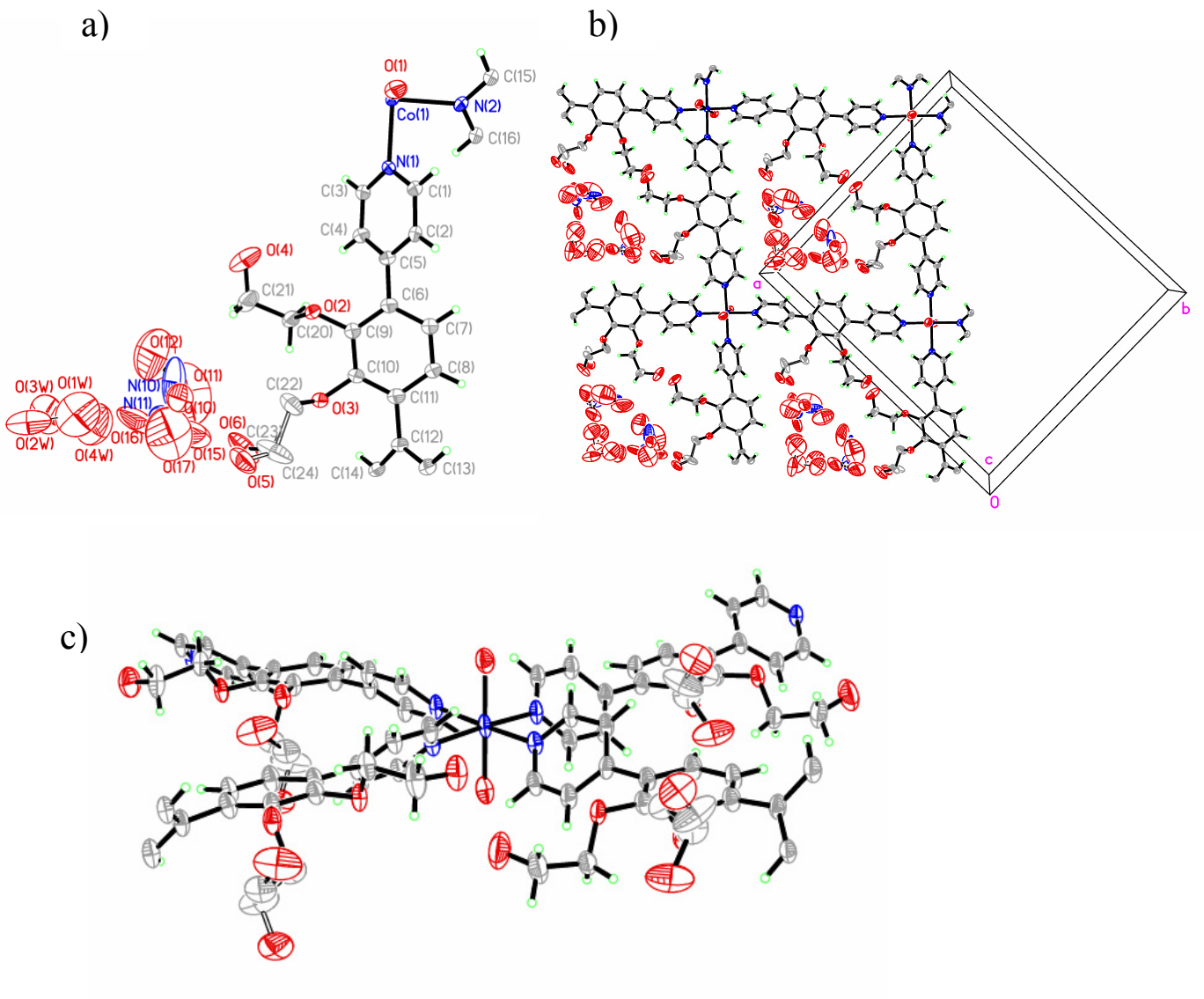

Figure S6. ORTEP view (50% probabilit y level) of **2'** (80 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Co(II) atom. The thermal temperature factors of all non-hydrogen atoms were anisotropically refined. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered ethylene glycol groups, water molecules and nitrate ions.

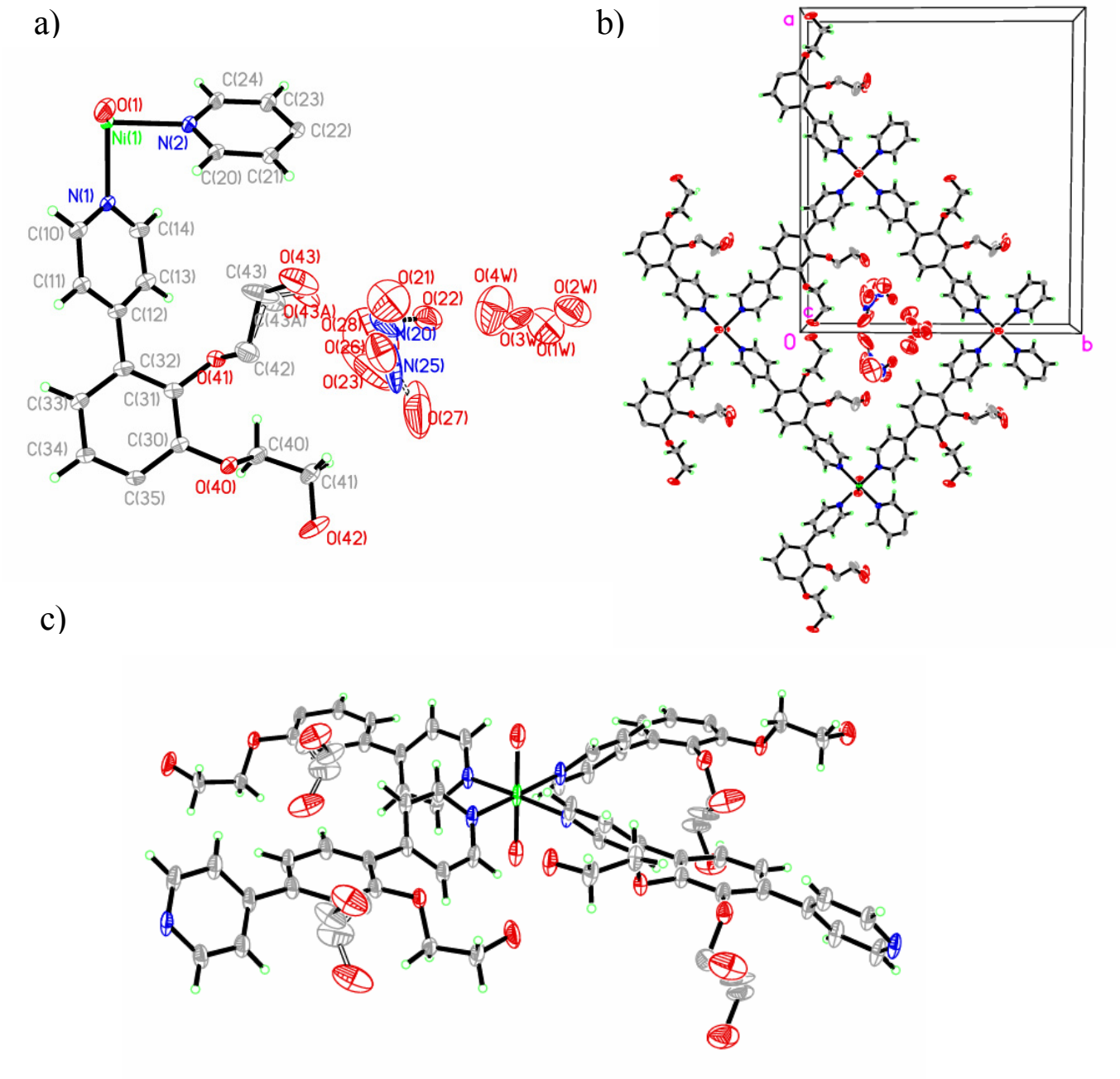

Figure S7. ORTEP view (50% probabilit y level) of **3** (80 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Ni(II) atom. The thermal temperature factors of all non-hydrogen atoms were anisotropically refined. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered ethylene glycol groups, nitrate ions and water molecules.

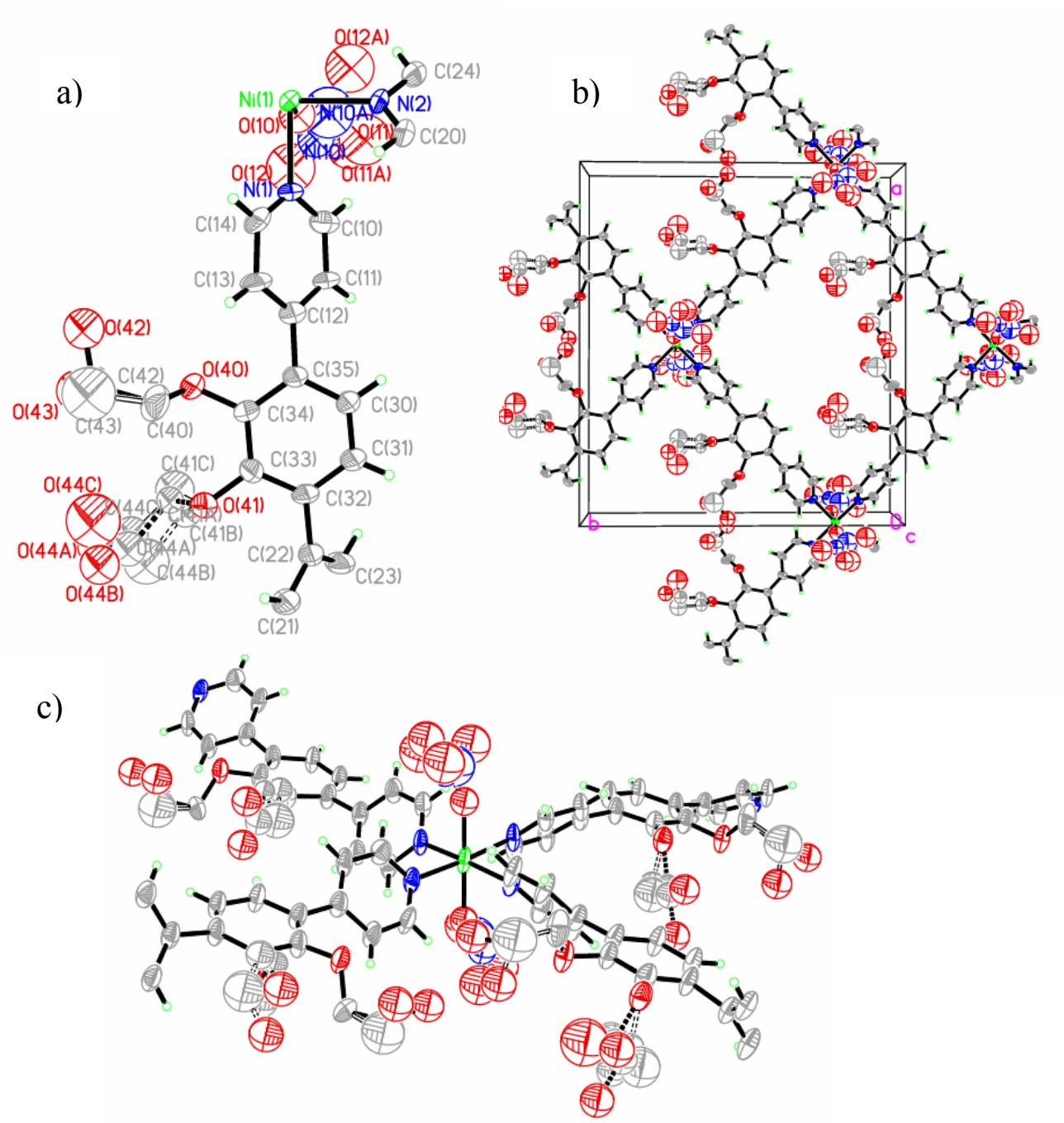

Figure S8. ORTEP view (50% probability level) of **7** (373 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Ni(II) atom. The thermal temperature factors of ethylene glycol groups, nitrate ions and guest molecules were isotropically refined because of severe disorder. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered nitrate ions and ethylene glycol groups.

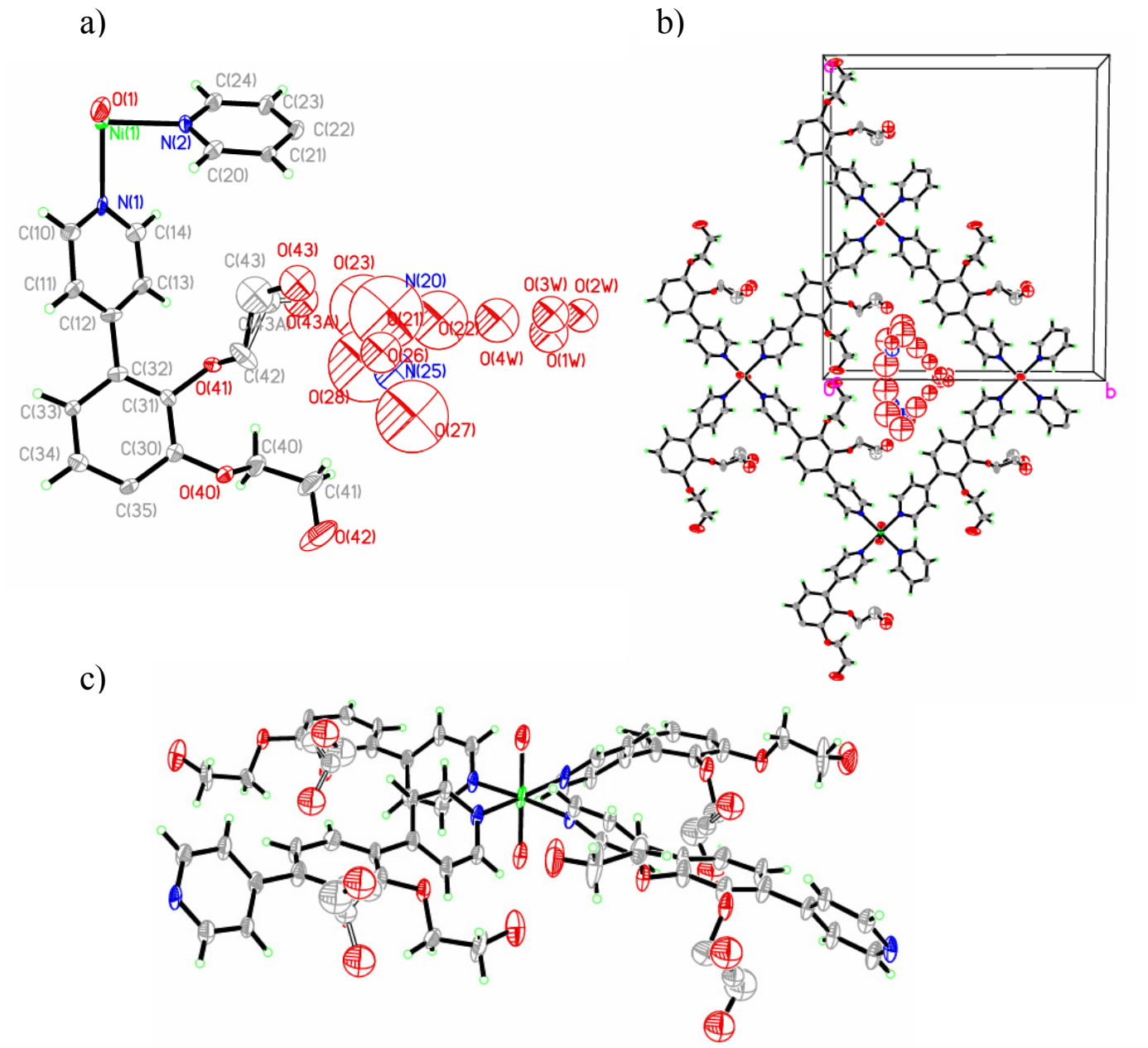

Figure S9. ORTEP view (50% probability level) of **3'** (80 K). (a) The crystallographic asymmetric unit. Hydrogen atoms are not labeled. (b) The 2-D layer. (c) The geometry around Ni(II) atom. The thermal temperature factors of ethylene glycol groups, nitrate ions and guest molecules were isotropically refined because of severe disorder. Hydrogen atoms were treated as a riding model. Several restraints were applied to disordered nitrate ions, ethylene glycol groups and water molecules.