Electronic Supporting information for "Step by Step Crystal-to-Crystal Transformation from 1D $K_2Cu(C_2O_4)_2(H_2O)_4$ (1) to 1D $K_2Cu(C_2O_4)_2(H_2O)_2$ (2), then 1D $K_2Cu(C_2O_4)_2$ (3) by Dehydration"

The IR experiment of single crystal was carried out on Bio-rad FTS6000/UMA500 Microscope with 4 cm^{-1} resolution at room-temperature.

Thermogravimetric analysis was carried out on a Shimadzu DTG-60 analyzer at a 10°C/min heating rate from room-temperature to 550°C. DSC experiment of **1** was carried out on TA Q600 and Diamond D800 with sample sealed in Al cap after crystal removed from solution as soon as possible.

Because **1** is unstable when exposure to air, the single crystal X-ray diffraction data was collected at 173 K. For compare, the single crystal X-ray diffraction data of **2** and **3** were collected at 173K

The single crystal X-ray diffraction data was collected at 173 K on Rigaku diffractometer with confused monochromated Mo Kα (λ = 0.71073 Å) radiation.[1] The structure was solved by direct method and refined by full-matrix least-squares on F^2 using SHELX program, with anisotropic thermal parameters for all non-hydrogen atoms.[2] Hydrogen atoms of H_2O were located by difference Fourier map and refined isotropically. The related crystallographic data was listed on Table S1.

The powder X-ray diffraction pattern was obtained on a Rigaku RINT2000 diffractometer at room temperature with Cu Ka (λ = 1.54056 Å) radiation in a flat-plat geometry.

Magnetization measurements were performed against tightly packed polycrystalline sample in capsule on a Quantum Design MPMS 7XL SQUID system. Susceptibility data were corrected for diamagnetism of sample by Pascal constants (-142.8×10^{-6} cm^3mol^{-1} for **1**; -117.8×10^{-6} cm^3mol^{-1} for **2**; -91.8×10^{-6} cm^3mol^{-1} for **3**) and background by experimental measurement on the sample holder.

References:

[1] Rigaku CrystalClear (2007). Version 1.4.0. Rigaku Inc., Tokyo, Japan.

[2] Sheldrick, G. M. (2008). Acta Cryst. A64, 112-122.

Table S1. Crystallographic data of 1-3

Compound	1	2	3
formula	C4H8CuK2O12	C4H4CuK2O10	C4CuK2O8
Color	Blue-purple	Blue	Blue
Fw	389.84	353.81	317.78
F(000)	390	350	310
T, K	173	173	173
crystal system	Monoclinic	Triclinic	Monoclinic
space group	$P\,2_1/n$	$P\,\bar{1}$	$P\,2_1/n$
Cell parameters	3.7345(7) 14.764(3) 10.742(2) 90 93.23(2) 90, 591.3(2)	6.904(2) 8.660(3) 8.929(3) 107.950(4) 99.637(4) 97.637(4) 490.9(3)	4.8886(11) 6.6448(15) 13.171(3) 90 96.305(5) 90 425.25(17)
Z	2	2	2
D_c, g/cm^3	2.190	2.394	2.482
μ (Mo K_α), mm^{-1}	2.611	3.118	3.568
crystal size, mm^3	0.280*0.104*0.094	0.293*0.252*0.198	0.12*0.08*0.07
T_{min} and T_{max}	0.6079, 0.8074	0.4632,1.0000	0.7782, 1.0000
θ_{min}, θ_{max} , °	3.350, 27.48	3.557, 27.54	3.112,27.468
Completeness, %	99.3	97.8	99.7
no. total reflns.	4206	4162	3644
no. uniq. reflns. (R_{int})	1341(0.0373)	2211(0.0316)	977(0.0472)
no. obs. [I≥2σ(I_0)]	1268	2158	916
no. params.	104	173	70
*R*1,*wR*2 [I ≥ 2σ (I_0)]	0.0304, 0.0623	0.0283, 0.0774	0.0332, 0.0753
*R*1,*wR*2 (all data)	0.0333, 0.0640	0.0290, 0.0781	0.0367, 0.0768
GOF	1.132	1.075	1.174
[a]Δρ, e/Å^3	0.421/-0.354	0.525/-0.556	0.433/-0.405
[b]Max. and mean Δ/σ	0.000/0.000	0.000/0.000	0.000/0.000
CCDC	851224	844292	844291

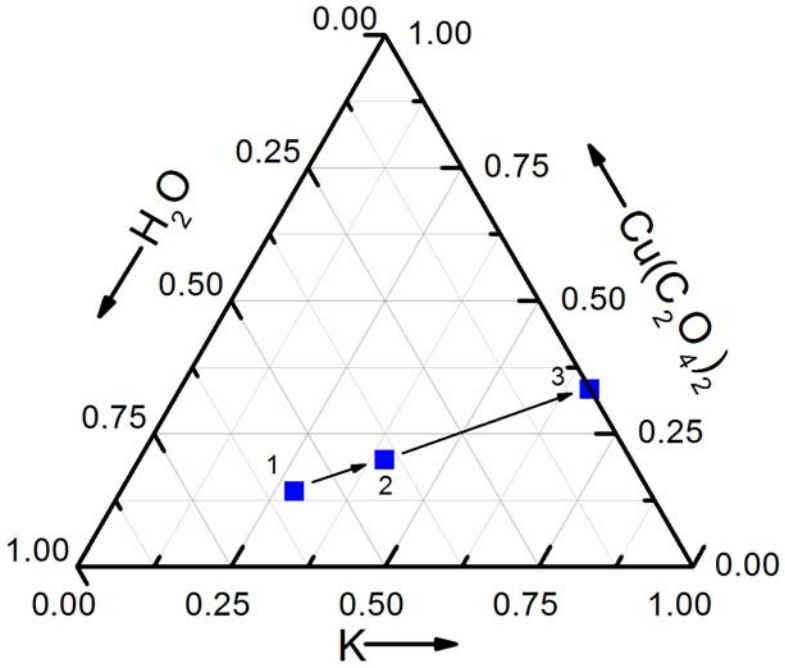


Figure S1. Phase-diagram of ternary K-$Cu(C_2O_4)_2$-H_2O system.

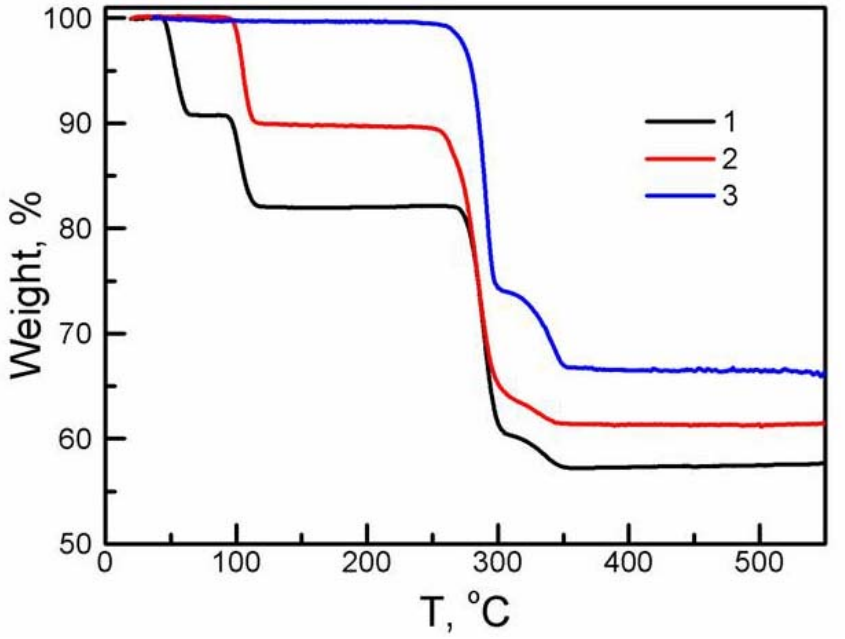


Figure S2. TGA plots of **1**-**3**.

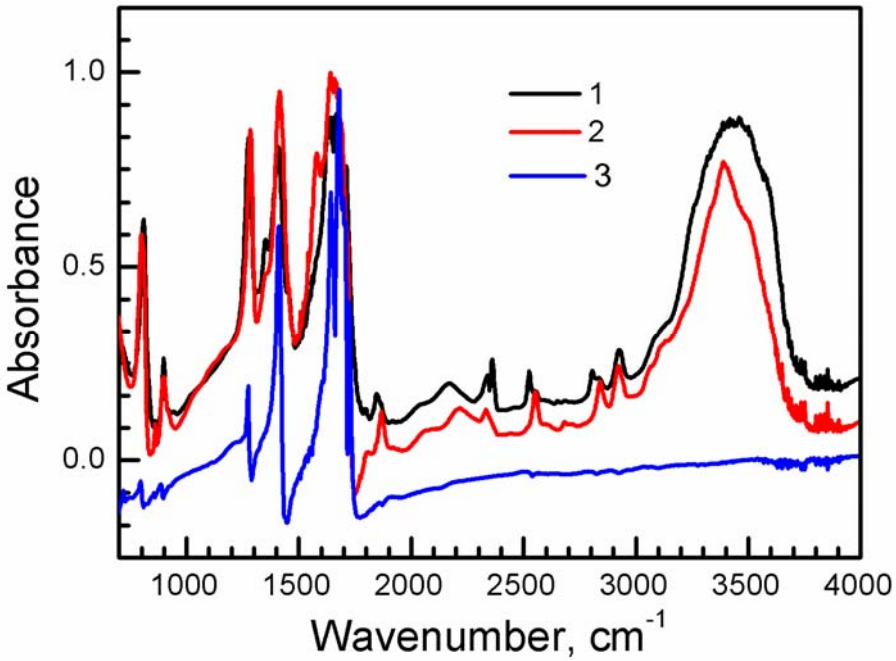


Figure S3. IR spectra on single crystal **1**, **2** and **3**.

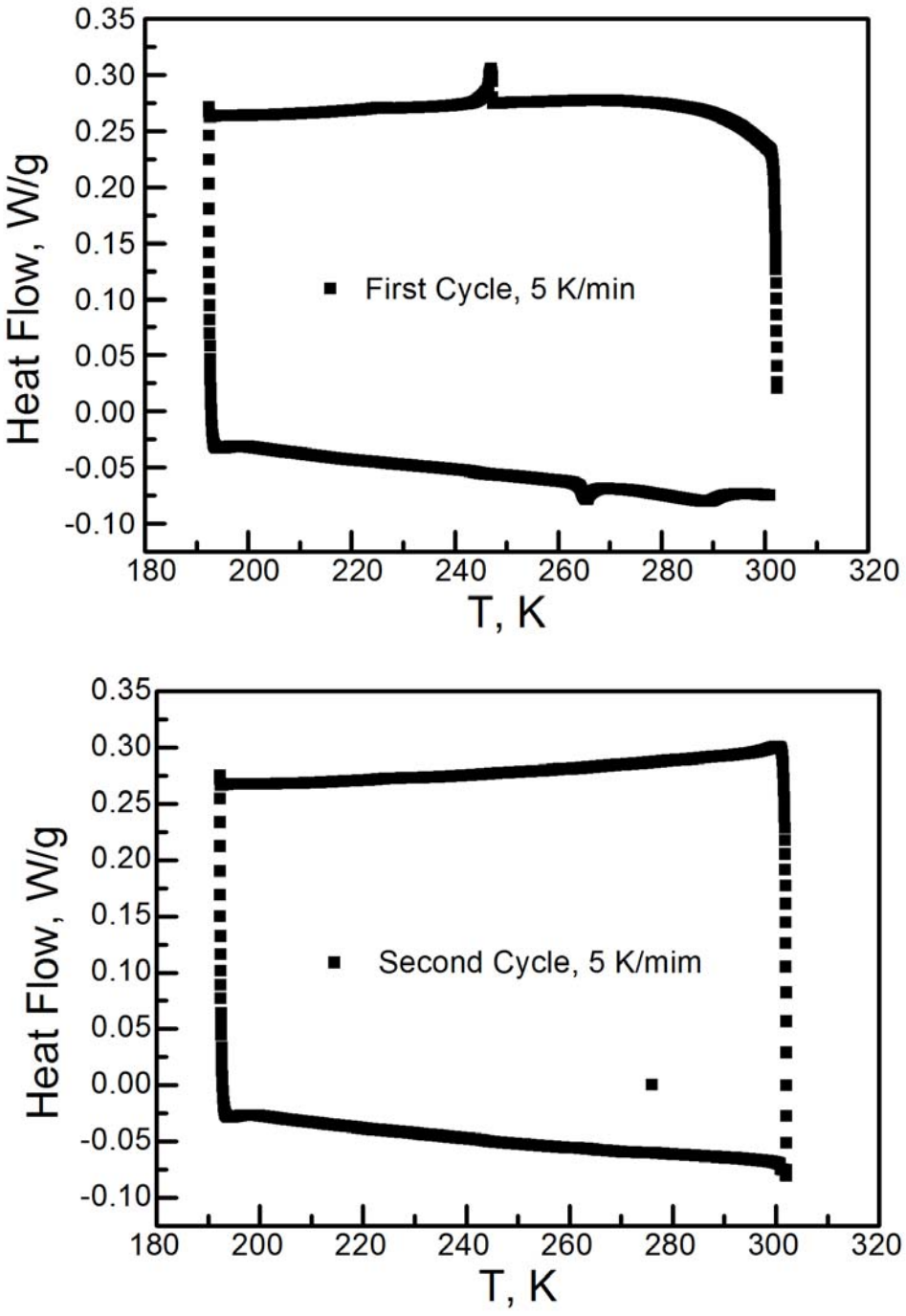


Figure S4. Differential Scanning Calorimeter of **1** with rate of 5 K/min between room-temperature and 190 K. The exotherm and endotherm which observed in first cycle measurement (top) were not observed in the second cycle measurement (down). The powder X-ray diffraction pattern after second cycle measurement is same as **2**. This means **1** transferred into **2** thoroughly.

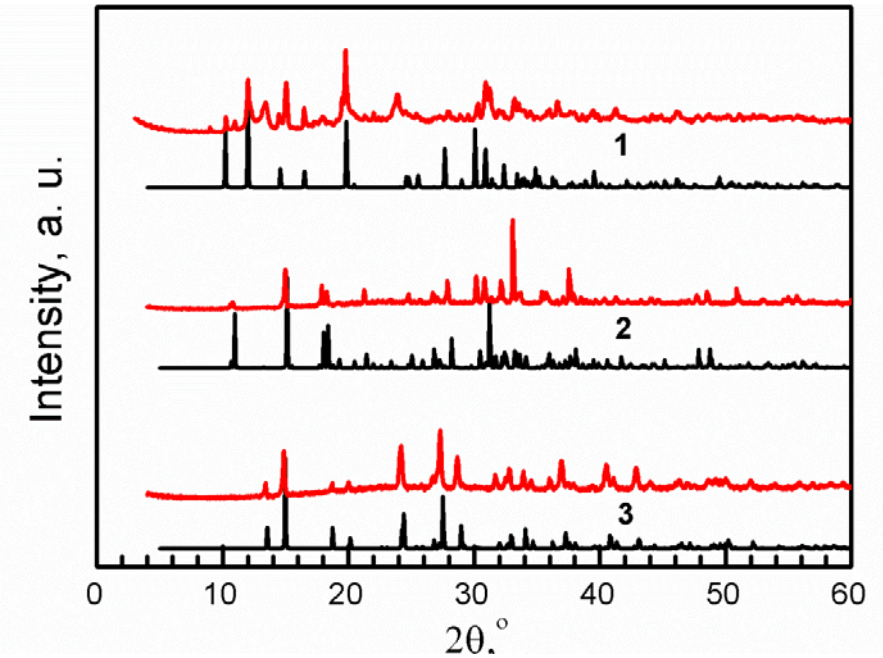
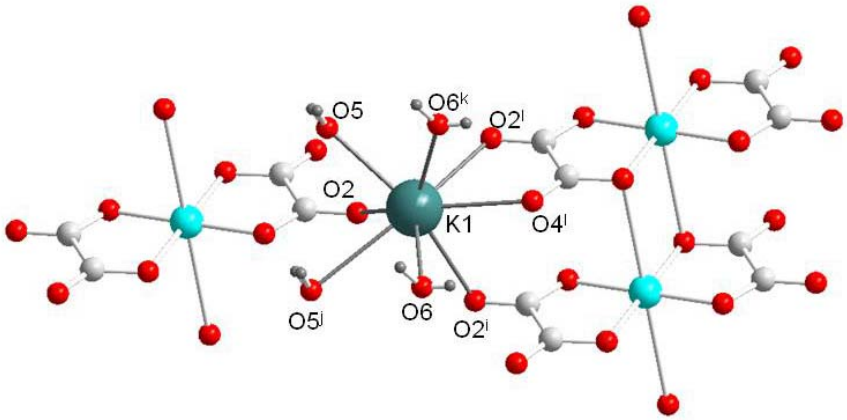
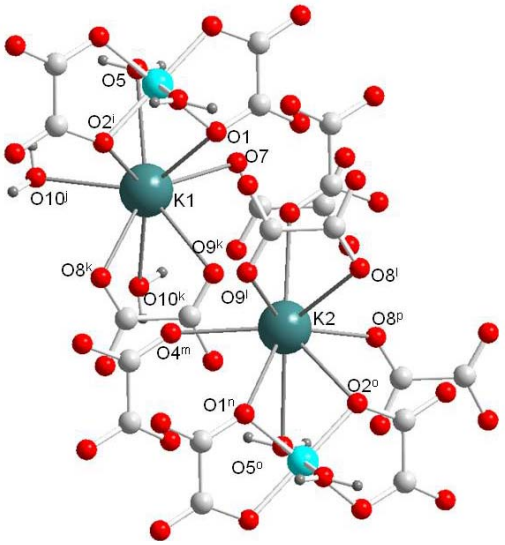


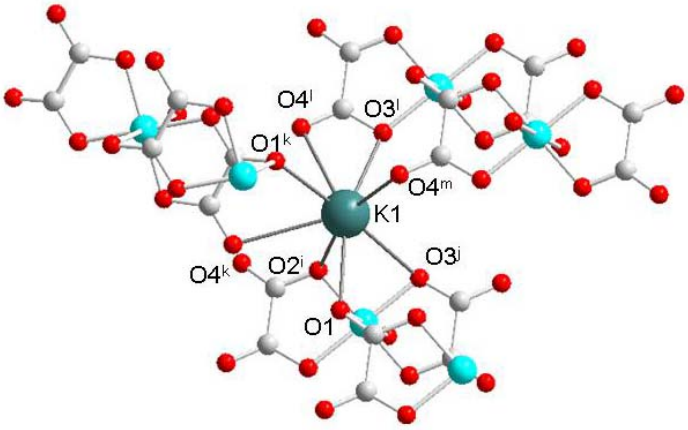
Figure S5. Powder diffraction pattern of compounds **1**, **2** and **3** at room-temperature. Black lines: simulated; red lines: experimental data. Although the experiment of **1** was carried out as soon as possible with crystal covered by Nephone-oil, there are still trace of **2** and **3** in experiment data **1**. The PXRD pattern of 2 and 3 are good.



1



2



3

Figure S6. Coordination configuration of K^+ in $K_2Cu(C_2O_4)_2(H_2O)_4$ (**1**, symmetry code: i: -x,-y,1-z; j: -1+x,y,z; k: 1+x,y,z; l: 1-x,-y,1-z), $K_2Cu(C_2O_4)_2(H_2O)_2$ (**2**, symmetry code: i: -x,-y,-z; j: -1+x,y,z; k: 1-x,1-y,1-z; l:x,y,-1+z; m: x,1+y,z; n:1-x,1-y,-z; o:1+x,1+y,z; p:2-x,1-y,1-z;) and $K_2Cu(C_2O_4)_2$ (**3**, symmetry code: i:1+x,y,z;j:1-x,1-y,1-z;k:1.5-x,0.5+y,0.5-z; l:1+x,1+y,z; m:x,1+y,z).

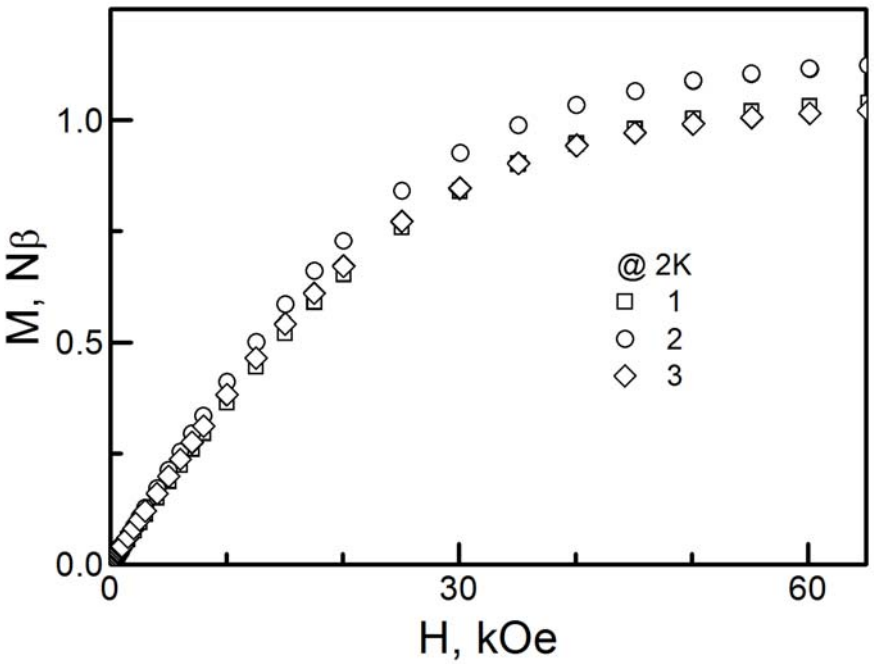


Figure S7. Isothermal magnetization at 2 K.