Supporting Information

Electron transfer induced magnetic ordering of metal-cyanide magnets

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Table S1. The stoichiometry *y* in all $Fe(TCNE)_y$ samples of different synthesis temperature of 368 K and 410 K.

368 K	**n (TCNE/mmol)**	**0.4**	**0.8**	**3.2**	
	y	0.19	0.88	1.00	
410 K	**n (TCNE/mmol)**	**0.8**	**1.6**	**3.2**	**6.4**
	y	1.02	1.67	3.75	5.59

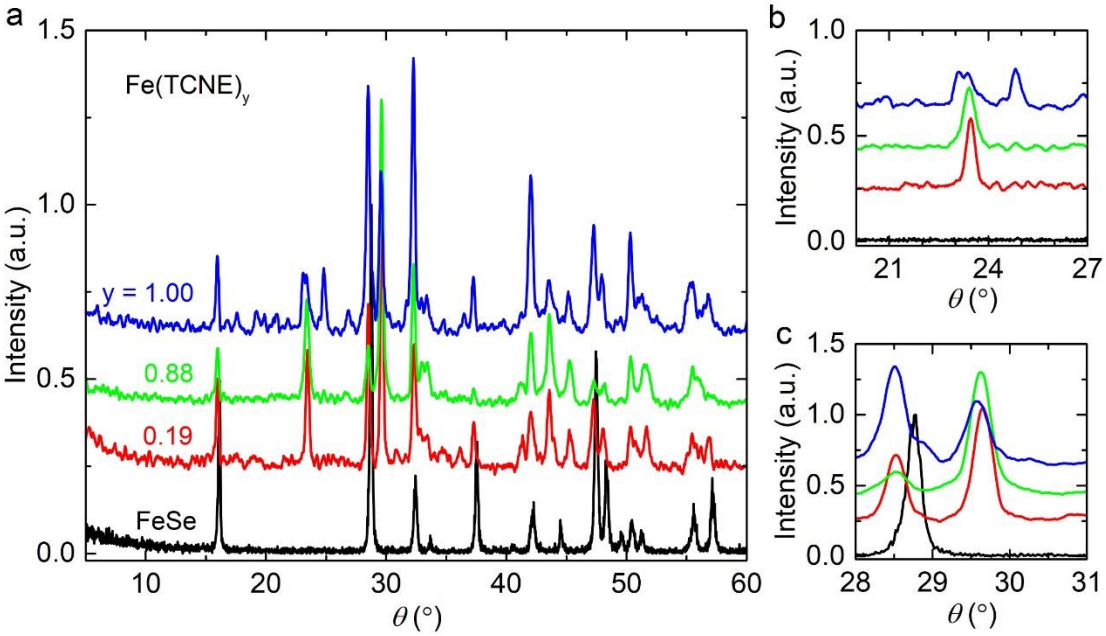

Figure S1. X-ray diffraction patterns (XRD) of Fe(TCNE)$_y$ synthesized at 368 K by FeSe template. (a) The whole XRD patterns from 5° to 60° show the structure evolution from FeSe to Fe(TCNE)$_y$ due to the Se substitution by TCNE. (b) New XRD peaks at around 24° of Fe(TCNE)$_y$ differ from that of FeSe. (c) Peak split at 29° identifies the new structure emerging during the substitution reaction.

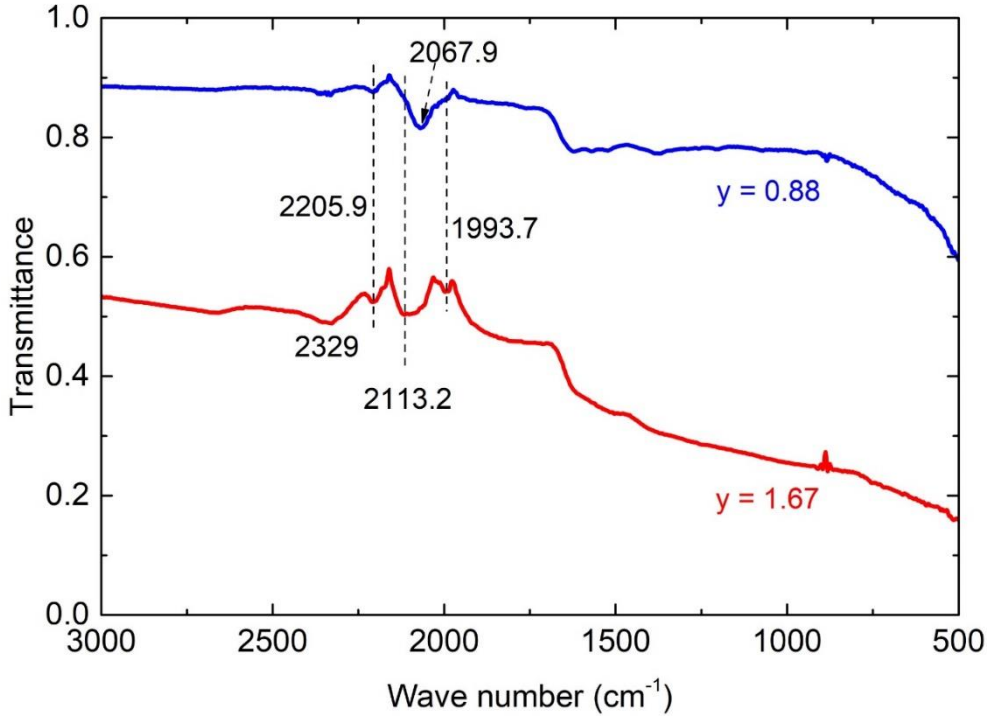

Figure S2. Infrared (IR) transmittance of Fe(TCNE)$_y$ (y =0. 88) at room temperature.

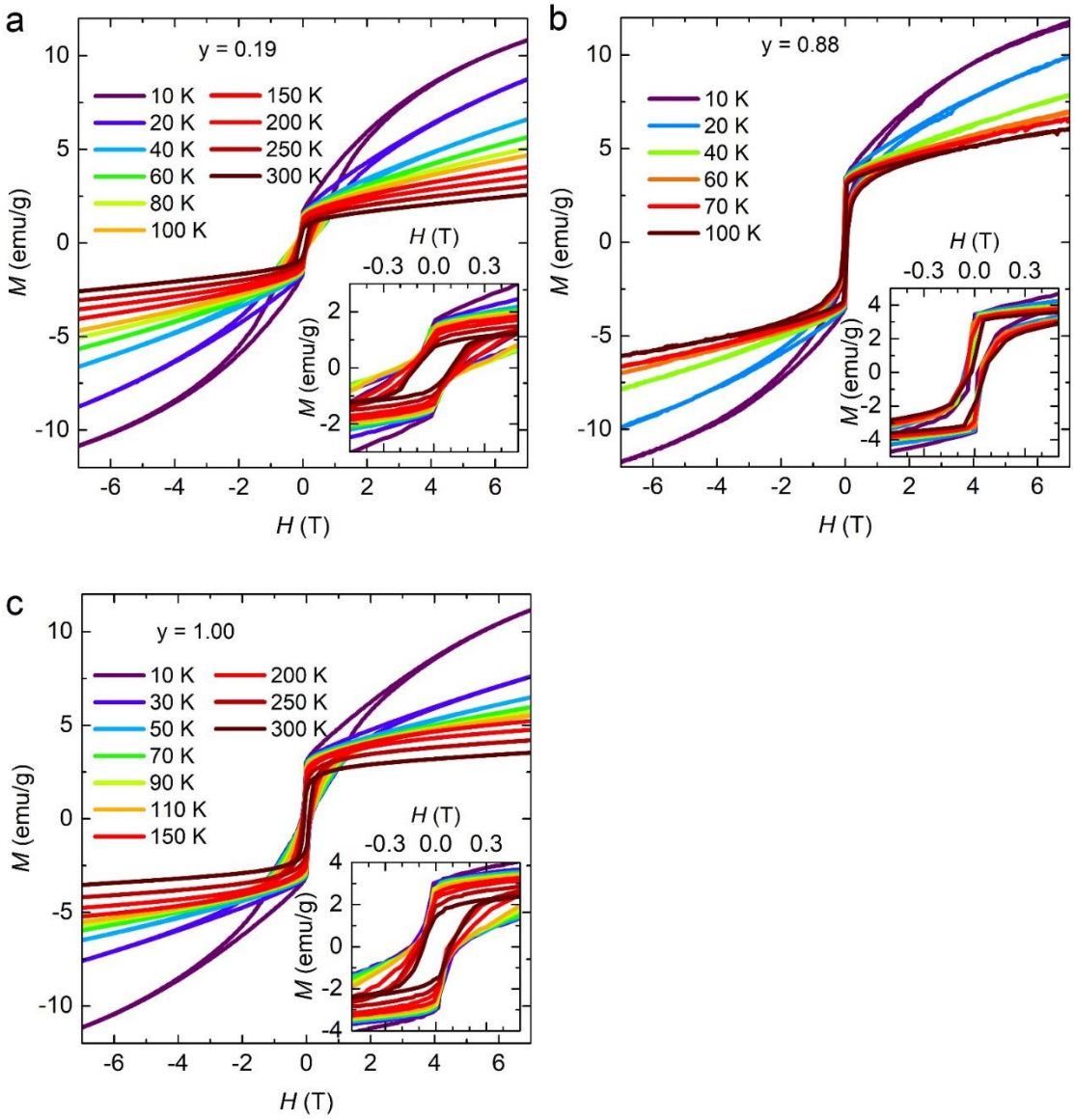

Figure S3. Magnetic field dependent of magnetization of $Fe(TCNE)_y$ (y = 0.19, 0.88 and 1.00) synthesized at 368 K.

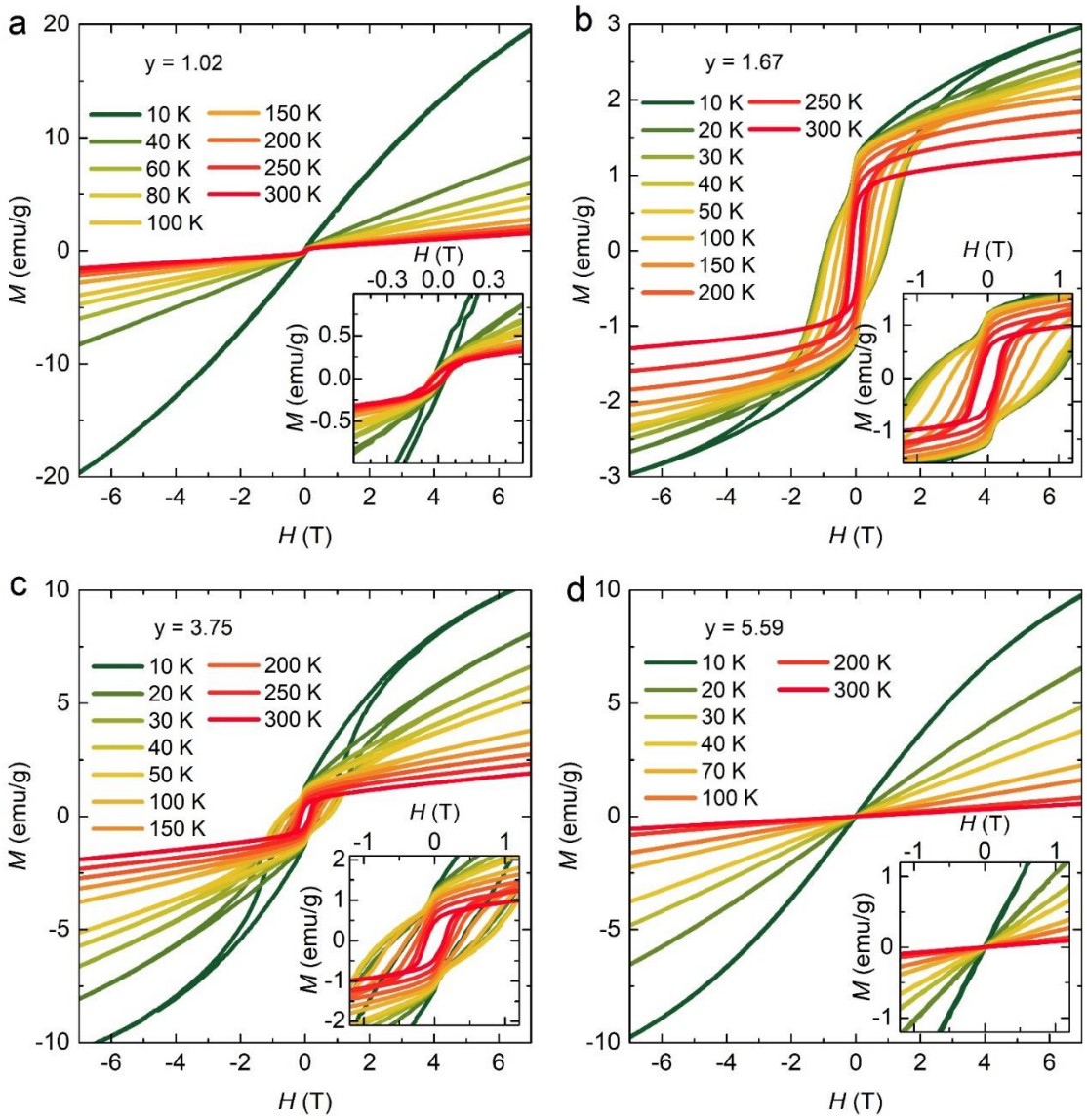

Figure S4. Magnetic field dependent of magnetization of $Fe(TCNE)_y$ (y = 1.02, 1.67, 3.75 and 5.59) synthesized at 410 K.

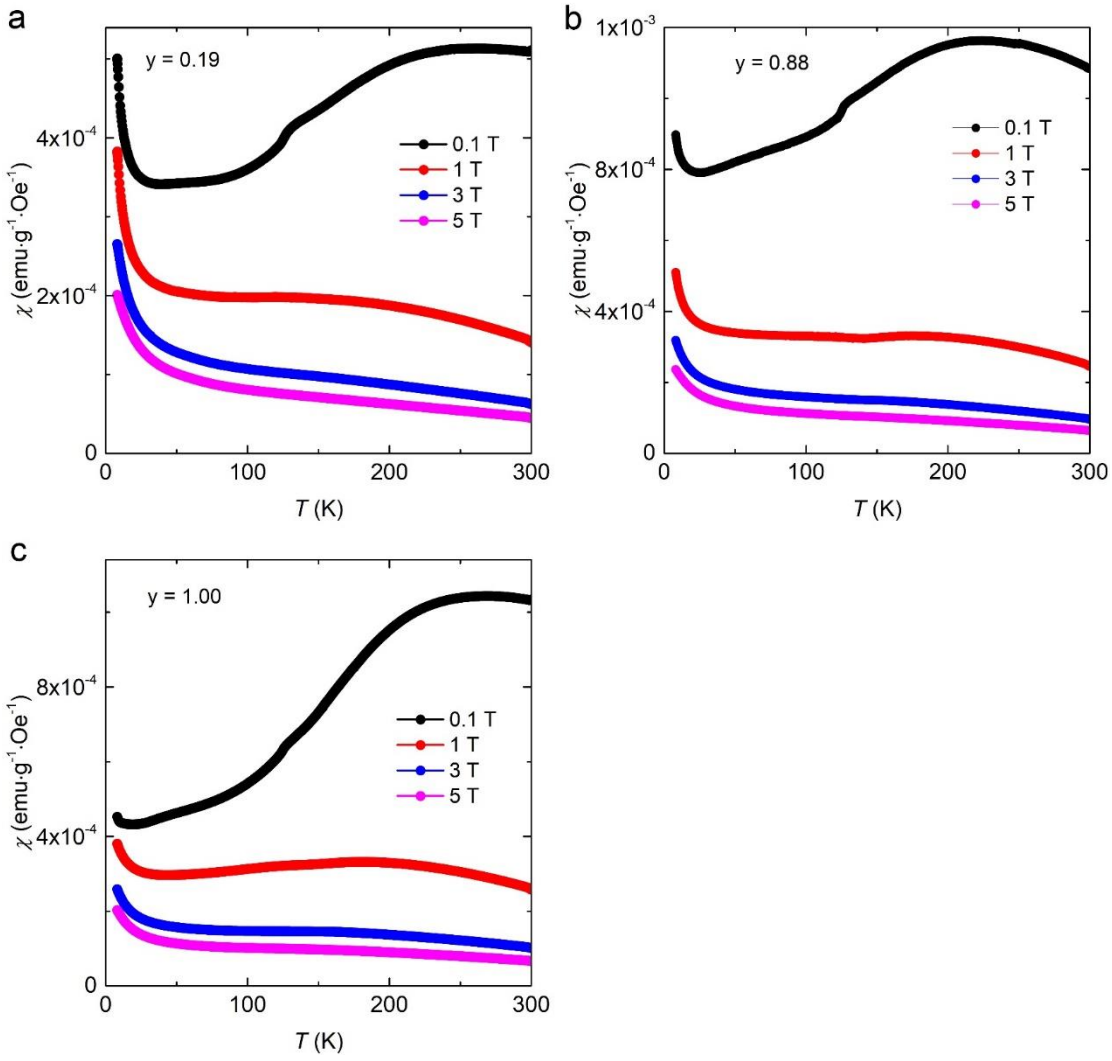

Figure S5. Temperature dependent of magnetic susceptibility of $Fe(TCNE)_y$ (y = 0.19, 0.88 and 1.00) synthesized at 368 K.

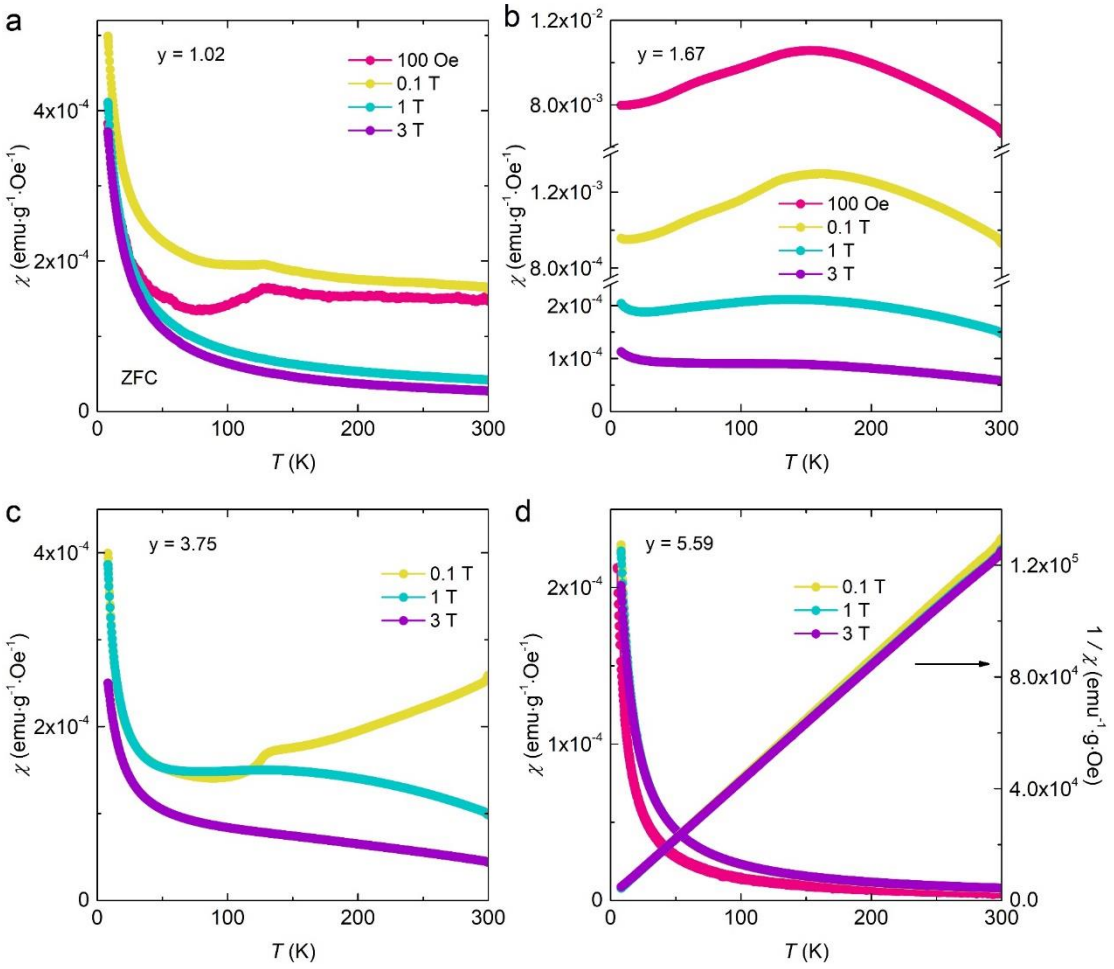

Figure S6. Temperature dependent of magnetic susceptibility of $Fe(TCNE)_y$ (y = 1.02, 1.67, 3.75 and 5.59) synthesized at 368 K.

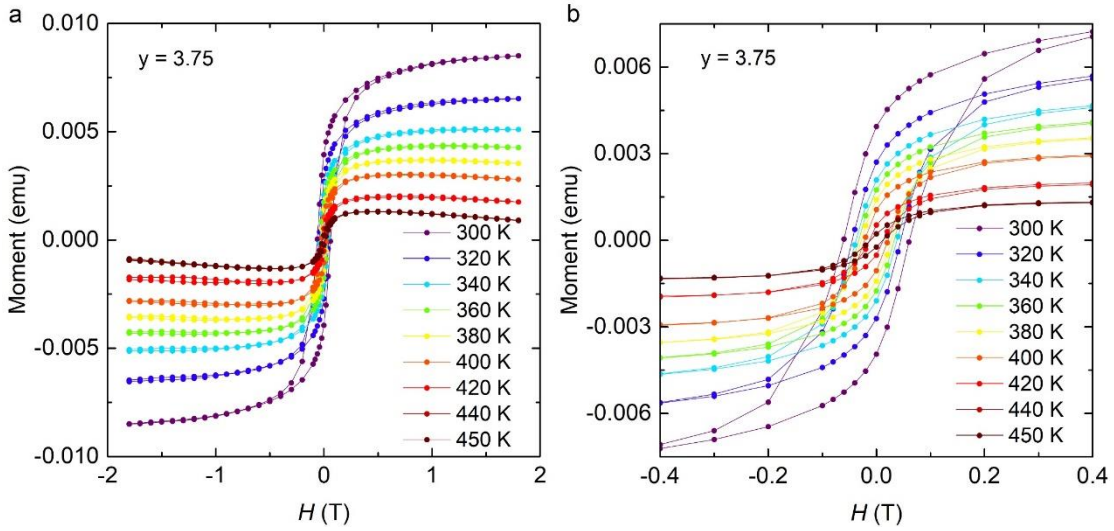

Figure S7. Magnetic field dependent of magnetization of $Fe(TCNE)_y$ (y = 3.75) at high temperature above 300 K.

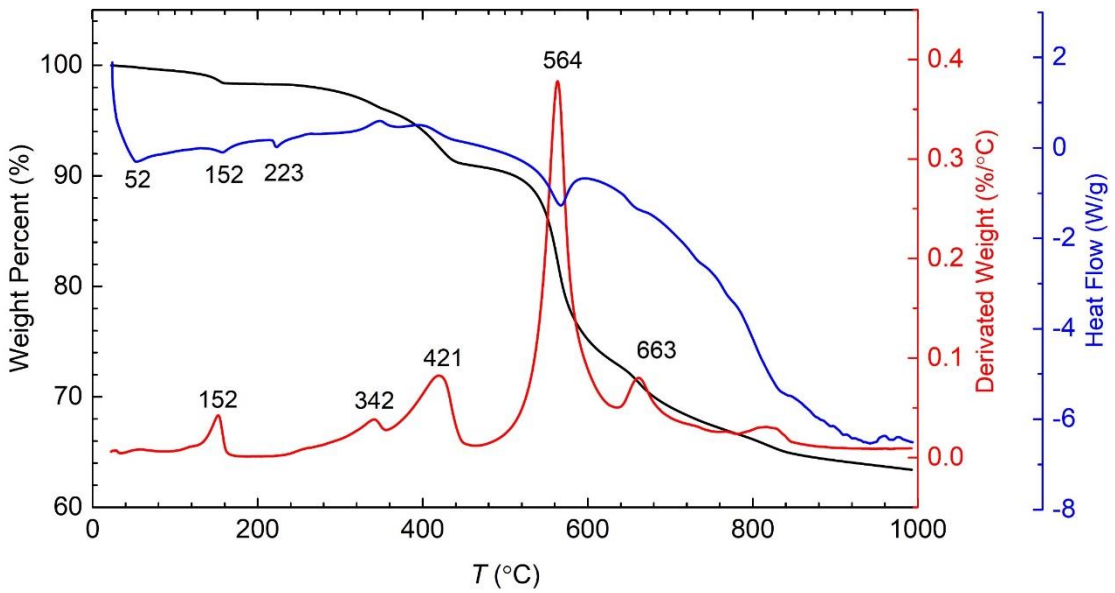

Figure S8. Thermal weigh and heat flow analysis of Fe(TCNE)$_y$.