

Supplementary Information

New layered organic-inorganic magnets incorporating azo dyes

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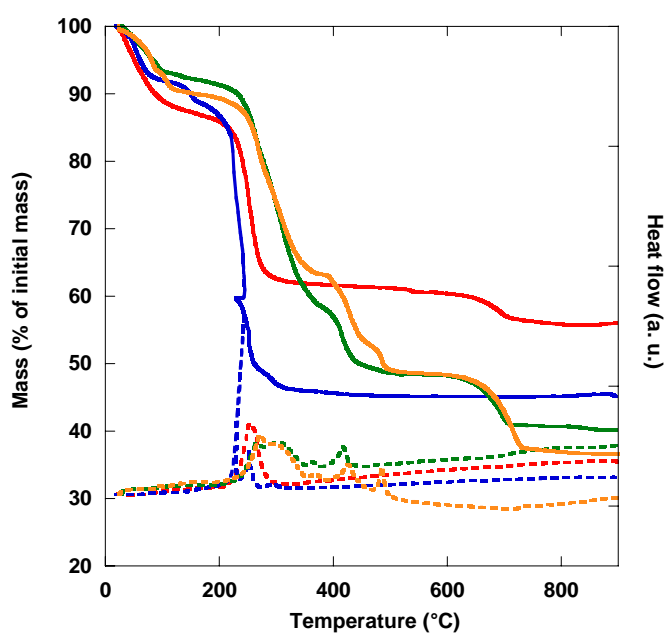


Fig. S1 TGA (full lines) and TDA (dotted lines) curves for **MRcCo** (1) (blue), **MOcCo** (2) (orange), **OrangeIVcCo** (3) (green), and **MY10cCo** (4) (red).

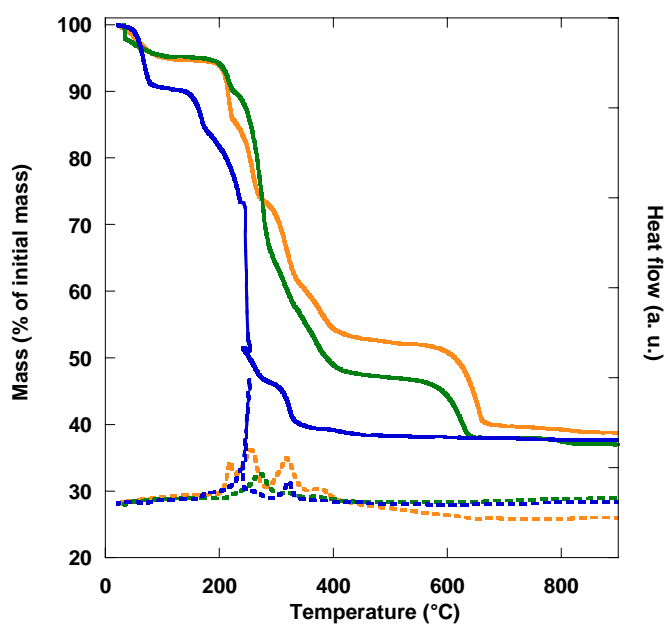


Fig. S2 TGA (full lines) and TDA (dotted lines) curves for **MRcCu** (5) (blue), **MOcCu** (6) (orange), and **OrangeIVcCu** (7) (green).

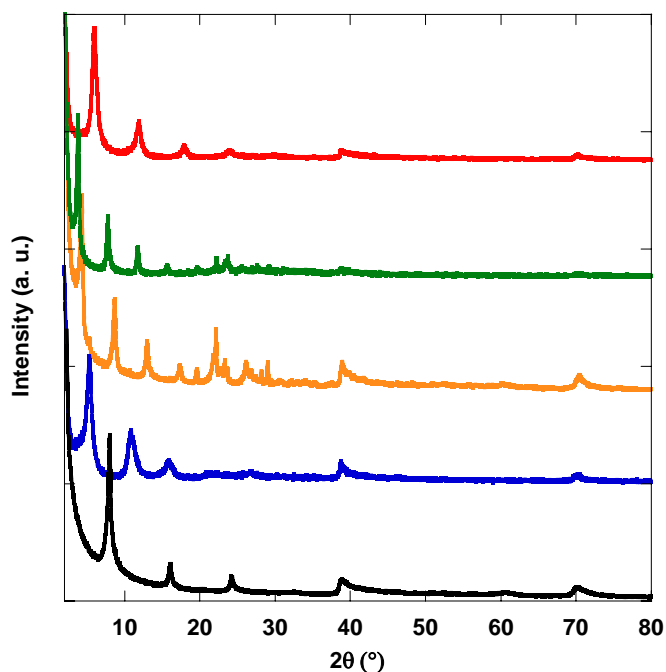


Fig. S3 Powder XRD patterns of $\text{Co}_2(\text{OH})_3(\text{OAc})\cdot\text{H}_2\text{O}$ (black), **MR-CO** (1) (blue), **MO-CO** (2) (orange), **OrangeIV-CO** (3) (green), and **MY10-CO** (4) (red) showing the shift of the $00l$ diffraction lines ($\text{Co K}\alpha_1 = 0.178897 \text{ nm}$).

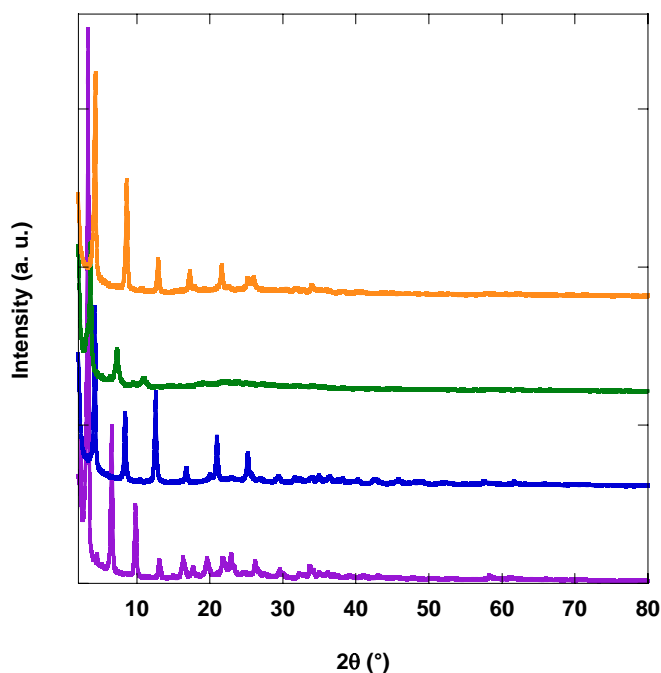


Fig. S4 Powder XRD patterns of $\text{Cu}_2(\text{OH})_3(\text{DS})$ (purple), **MR-CU** (5) (blue), **MO-CU** (6) (orange), and **OrangeIV-CU** (7) (green) showing the shift of the $00l$ diffraction lines ($\text{Cu K}\alpha_1 = 0.1540598 \text{ nm}$).

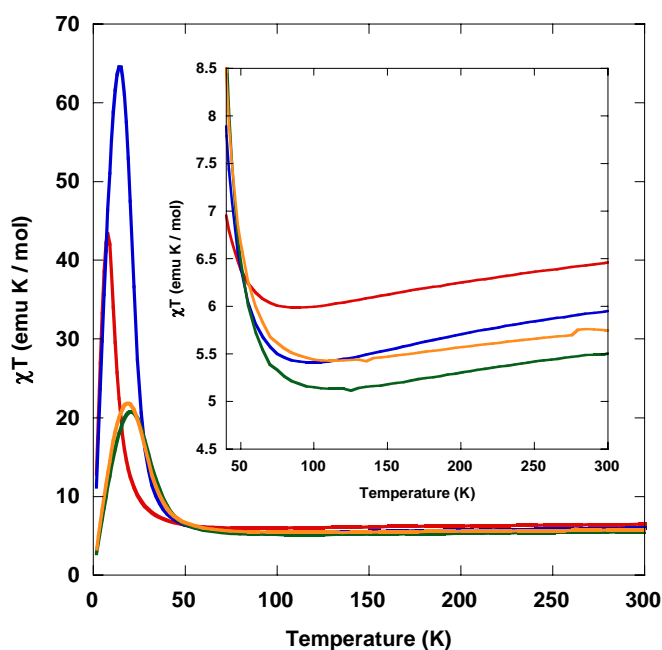


Fig. S6 Magnetic behaviour as χT vs T plots of compounds **MRcCo** (1) (blue), **MOcCo** (2) (orange), **OrangeIVcCo** (3) (green), and **MY10cCo** (4) (red).

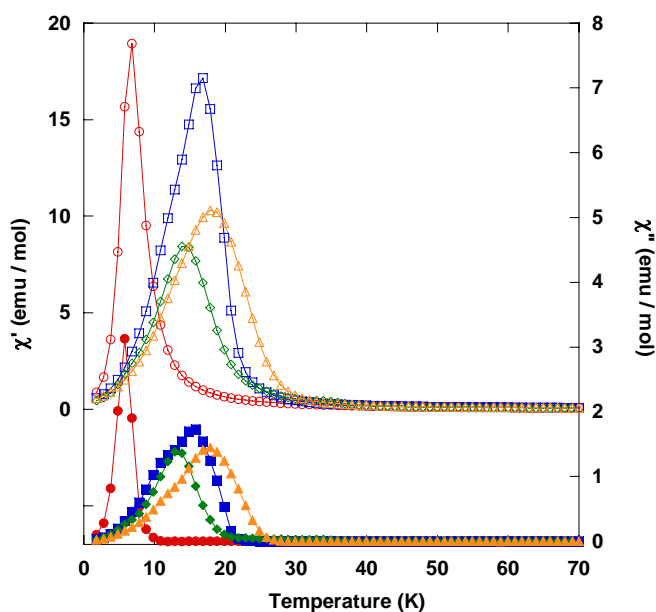


Fig. S7 In phase (χ') and out of phase (χ'') magnetic susceptibilities measured in an ac field of 0.35 mT at 100 Hz for **MRcCo** (1) (open and closed blue squares), **MOcCo** (2) (open and closed orange triangles), **OrangeIVcCo** (3) (open and closed green lozenges), and **MY10cCo** (4) (open and closed red circles) (full lines are just a guide for the eye).

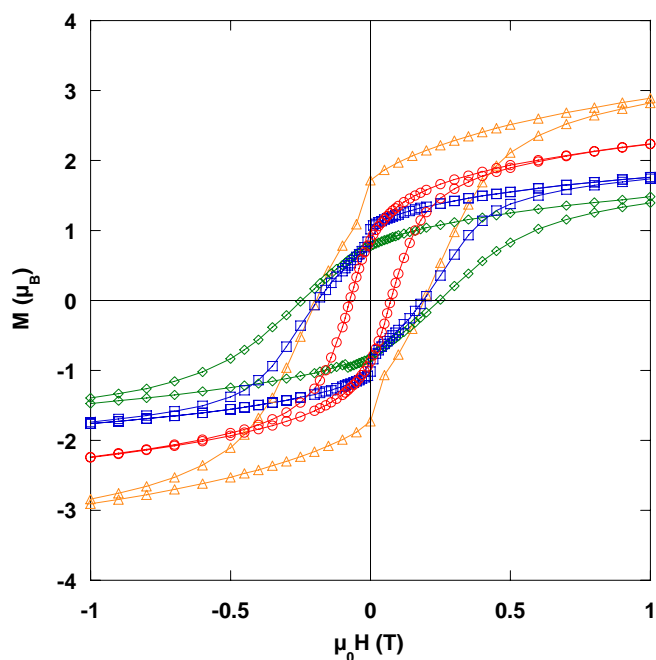


Fig. S8 Field dependence at $T = 1.8$ K of the magnetization of compounds **MRCo** (1) (open blue squares), **MOCo** (2) (open orange triangles), **OrangeIVCo** (3) (open green lozenges), and **MY10Co** (4) (open red circles) (full lines are just a guide for the eye).

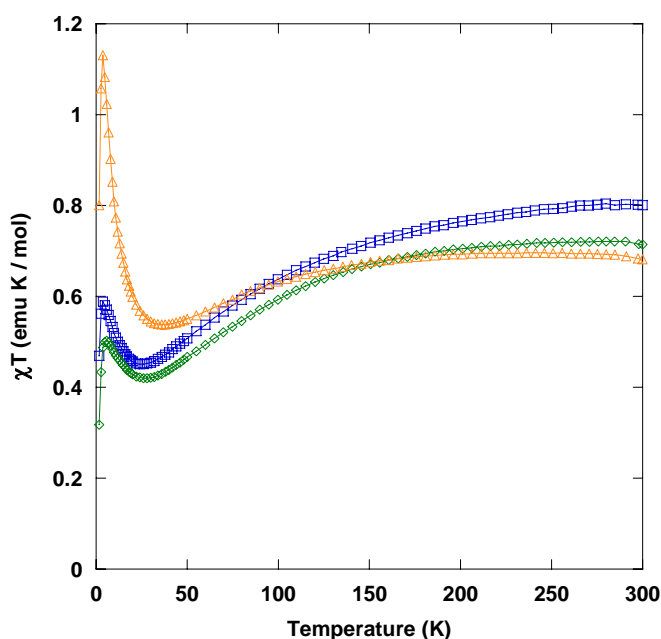


Fig. S9 Magnetic behaviour as χT vs T plots of compounds **MRCo** (5) (open blue squares), **MOCo** (6) (open orange triangles), **OrangeIVCo** (7) (open green lozenges) (full lines are just a guide for the eye).

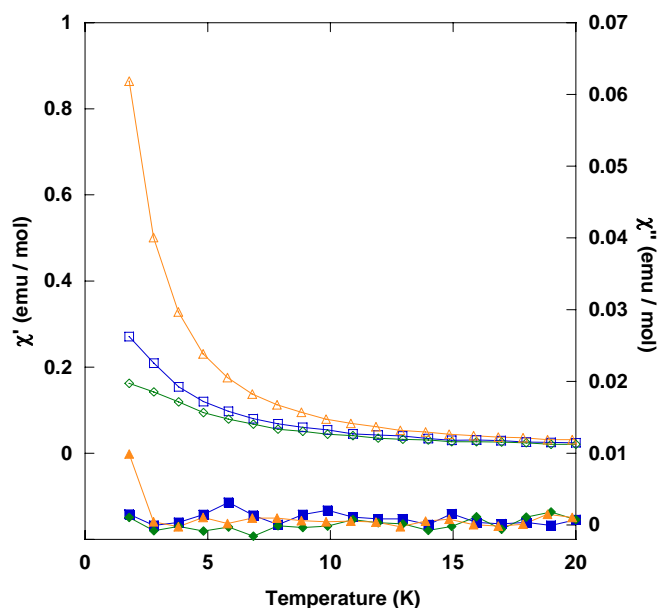


Fig. S10 Field dependence at $T = 1.8$ K of the magnetization of compounds **MR-Cu** (**5**) (open blue squares), **MO-Cu** (**6**) (open orange triangles), **OrangeIV-Cu** (**7**) (open green lozenges) (full lines are just a guide for the eye).

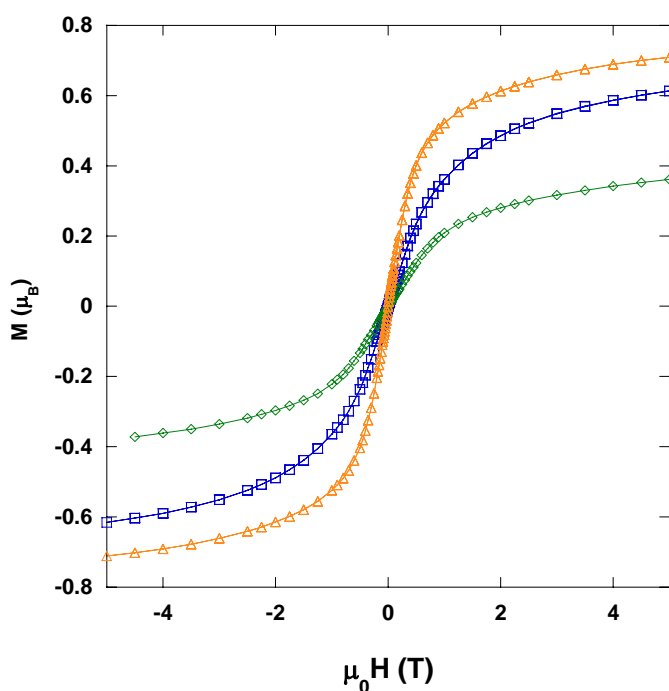


Fig. S11 In phase (χ') and out of phase (χ'') magnetic susceptibilities measured in an ac field of 0.35 mT at 100 Hz for **MR-Cu** (**5**) (open and closed blue squares), **MO-Cu** (**6**) (open and closed orange triangles), **OrangeIV-Cu** (**7**) (open and closed green lozenges) (full lines are just a guide for the eye).

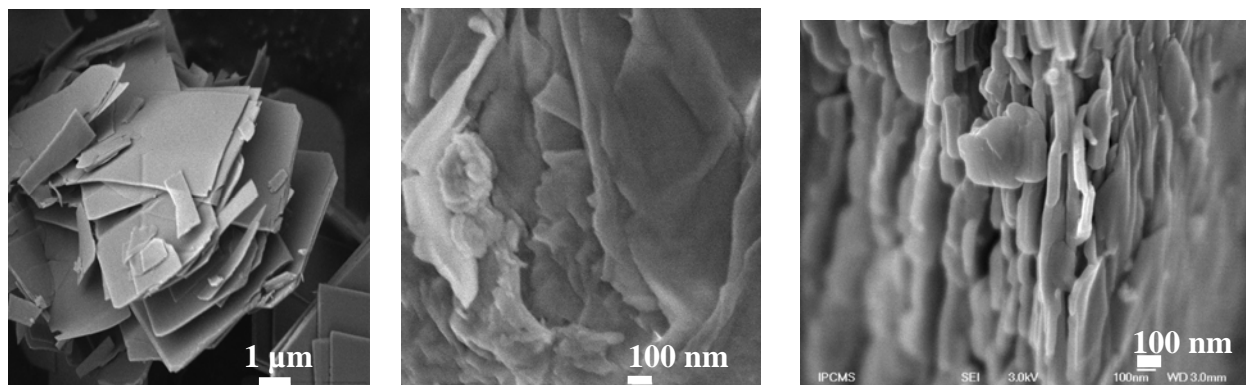


Fig. S12 SEM images of the starting compounds $\text{Cu}_2(\text{OH})_3(\text{Ac})\cdot\text{H}_2\text{O}$ (left), $\text{Co}_2(\text{OH})_3(\text{OAc})\cdot\text{H}_2\text{O}$ (middle), and $\text{Cu}_2(\text{OH})_3(\text{DS})$ (right).

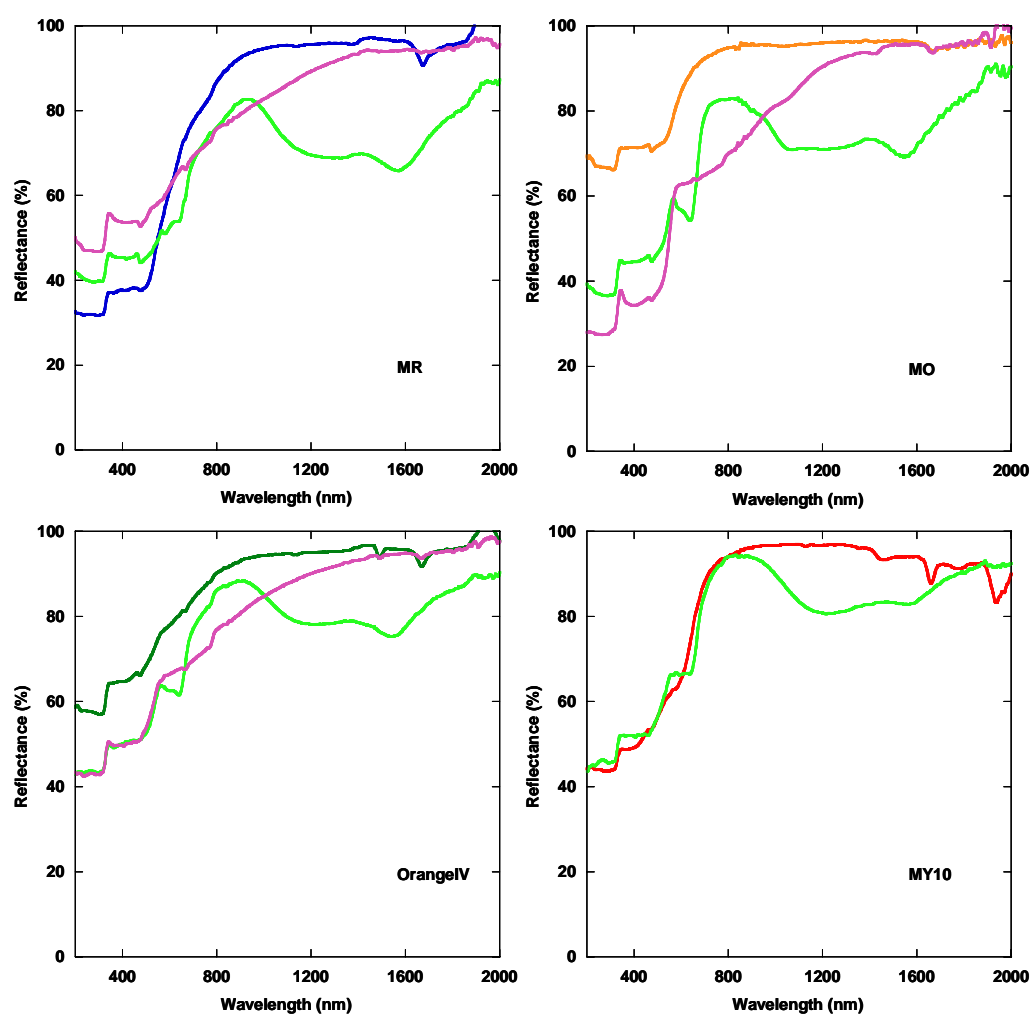
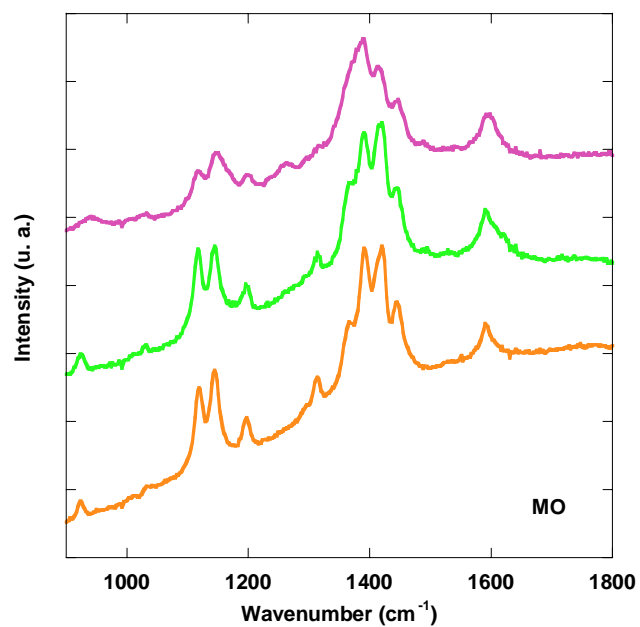
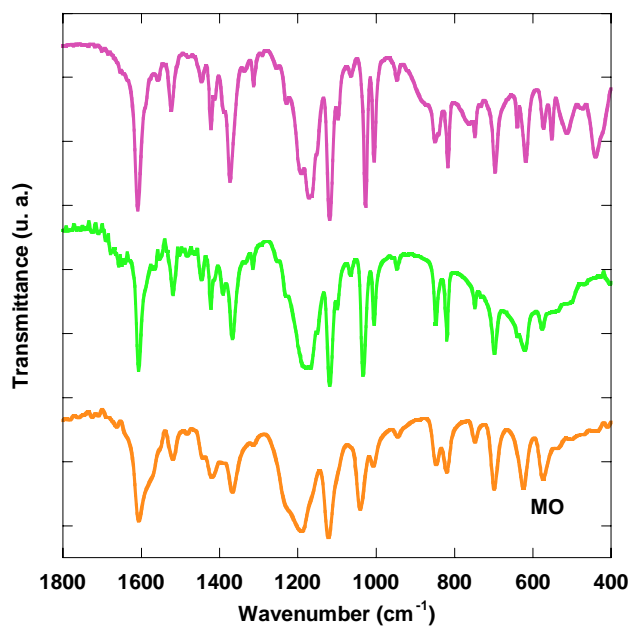
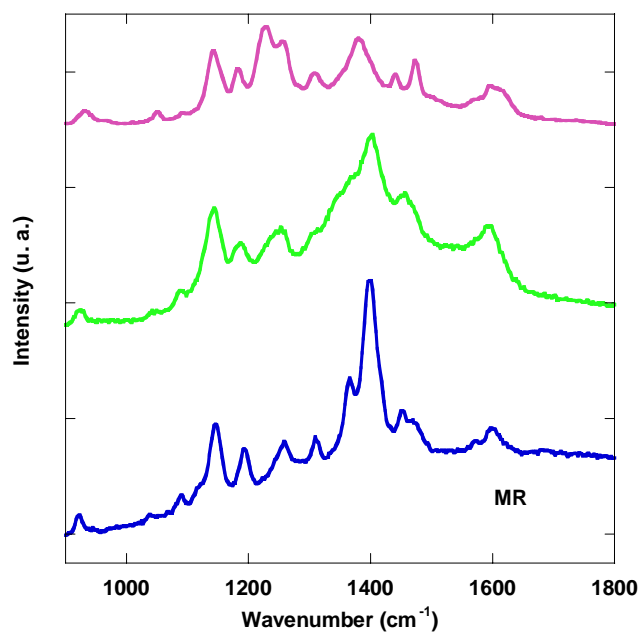
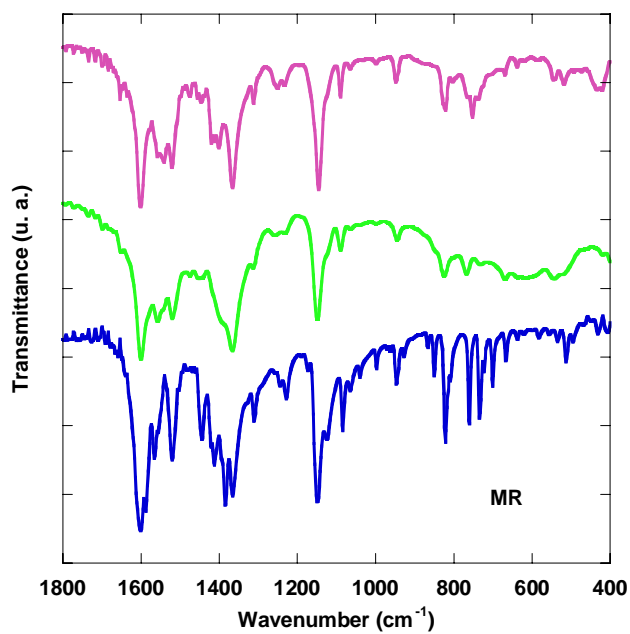


Fig. S13 UV-Vis. reflectance spectra of hybrid compounds containing Co (bright green) and Cu (purple), compared to the starting dyes **MR** (blue), **MO** (orange), **OrangeIV** (green), and **MY10** (red).



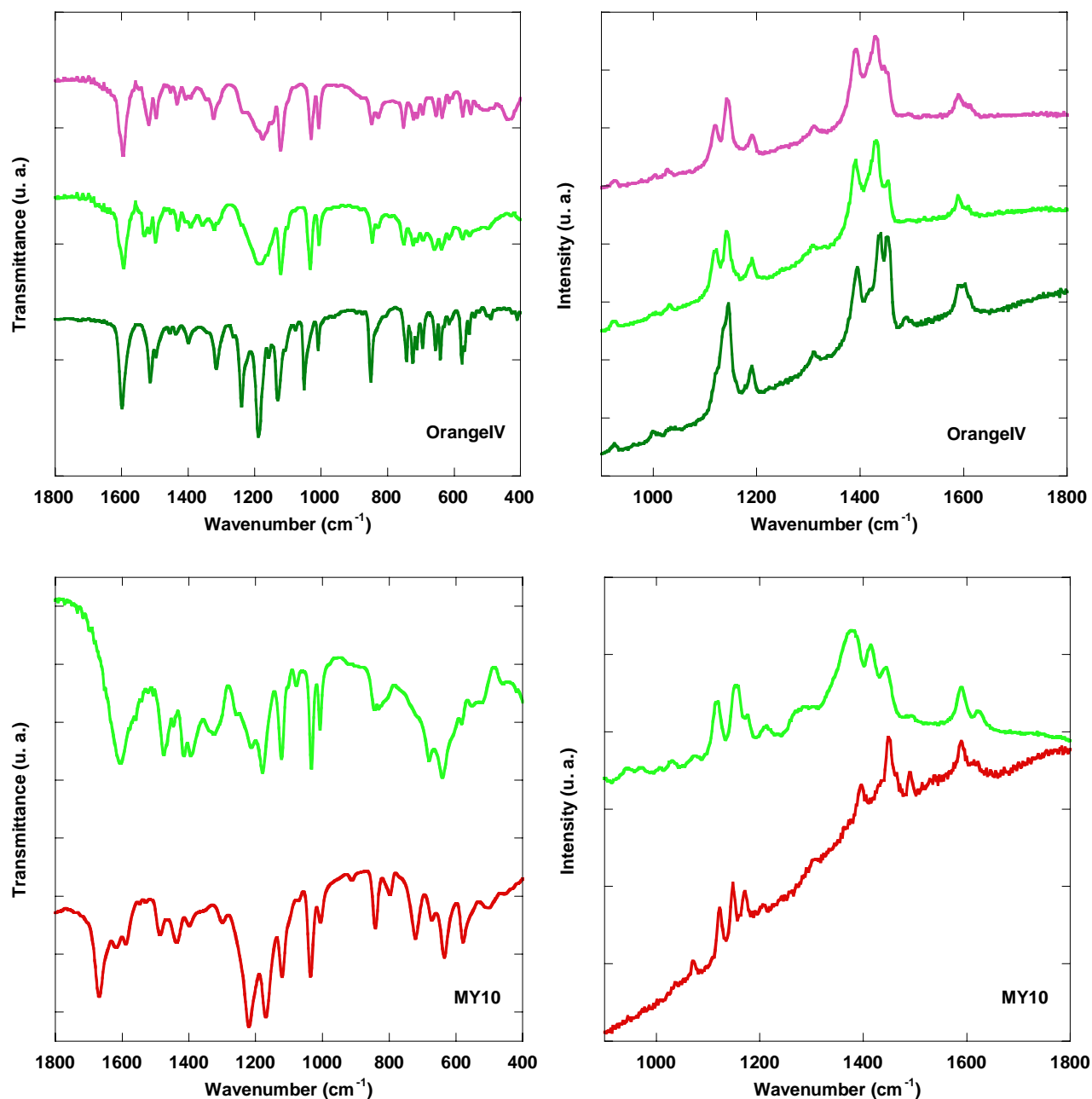


Fig. S14 IR (left) and Raman (right) ($\lambda_{\text{exc}} = 514.5 \text{ nm}$) spectra of the hybrid compounds containing Co (bright green) and Cu (purple), compared to the starting dyes **MR** (blue), **MO** (orange), **OrangeIV** (green), and **MY10** (red) (from top to bottom).