Supporting Information

Fig. S1  Complexation of Cu(II) salts with different arginine Schiff bases used in this study.
**Fig. S2** Superimposed images of (left) 1a and 1b (shown in green and blue respectively); (right) 2a and 2b (shown in green and blue respectively). The arginine side chains in 1 are obviously much deviated than that in 2.

**Fig. S3** Superimposed images of 3 (orange) with 1a (left) and 1b (right) respectively.
Fig. S4  Superimposed images of 3 (orange) with 2a (left) and 2b (right) respectively.

Fig. S5  CD spectrum of HRNap and HRMe ligands in MeOH.
Fig. S6  CD spectrum of 1, 2 and 3 in ethanol/ water mixture (1:1, v/v).
Fig. S7 $^1$H NMR of HRNap ligand.
Fig. S8 $^{13}$C NMR of HRNap ligand.
Fig. S9  COSY NMR of HRNap ligand.
Fig. S10 $^{1}$H-$^{13}$C HMQC NMR of HRNap ligand.
Fig. S11 $^1$H NMR of HRNO$_2$ ligand.
Fig. S12  COSY NMR of HRNO$_2$ ligand.
Fig. S13  $^1$H NMR of HRMe ligand.
Fig. S14  $^{13}$C NMR of HRMe ligand.
Fig. S15  COSY NMR of HRMe ligand.
Fig. S16  $^1$H-$^{13}$C HMQC NMR of HRMe ligand.
Fig. S17  TGA of [Cu(RNap)(OAc)]·2H₂O, 1.
*Fig. S18*  UV-visible spectrum of [Cu(RNap)(OAc)]·2H₂O, 1.
Fig. S19  Postitive mode ESI spectrum of [Cu(RNap)(OAc)]·2H₂O, 1 and its simulated isotopic spectrum.
Loss of 3 H₂O, Calc: 5.3%  
Loss of 3 H₂O, Calc: 5.3%  
6.864% (0.7744mg)  
4.311% (0.4364mg)  
70.00°C  
120.00°C  
225.00°C  

Fig. S20  TGA of [Cu(RNO₂)Cl(H₂O)]·[[Cu(RNO₂)(H₂O)₂](ClO₄)]·3H₂O, 2.
Fig. S21  UV-visible spectrum of $[\text{Cu(RNO}_2\text{)Cl(H}_2\text{O)}] \cdot [\text{[Cu(RNO}_2\text{(H}_2\text{O)}_2\text{)(ClO}_4)]} \cdot 3\text{H}_2\text{O}$, 2.
Fig. S22  Positive mode ESI spectrum of

\[ \text{[Cu(RNO}_2\text{)Cl(H}_2\text{O)}\cdot\text{[Cu(RNO}_2\text{(H}_2\text{O)}\text{)(ClO}_4\text{)}\cdot\text{3H}_2\text{O, 2}} \]

and its simulated isotopic spectrum.
Fig. S23  TGA of \([\text{Cu(RMe)(OAc)\cdot5H}_2\text{O, 3.}}\)
Fig. S24  UV-visible spectrum of $[\text{Cu(RMe)(OAc)}] \cdot 5\text{H}_2\text{O}$, 3.
Fig. S25  Postitive mode ESI spectrum of [Cu(RMe)(OAc)]·5H₂O, 3 and its simulated isotopic spectrum.